The PHILOSOPHICAL MAGAZINE

FIRST PUBLISHED IN 1798

UNIVERSITY OF HAWAII
LIBRARY
JUN 8 '54

L. 45 SEVENTH SERIES No. 363 April 1954

A Journal of Theoretical Experimental and Applied Physics

EDITOR
PROFESSOR N. F. MOTT, M.A., D.Sc., F.R.S.

EDITORIAL BOARD
SIR LAWRENCE BRAGG, O.B.E., M.C., M.A., D.Sc., F.R.S.
SIR GEORGE THOMSON, M.A., D.Sc., F.R.S.
PROFESSOR A. M. TYNDALL, C.B.E., D.Sc., F.R.S.

PRICE 15s. 0d.
Annual Subscription £8 0s. 0d. payable in advance

ED AND PUBLISHED BY TAYLOR & FRANCIS LTD., RED LION COURT, FLEET ST., LONDON, E.C.4.

Commemoration Number

To mark the 150th Anniversary of the

PHILOSOPHICAL MAGAZINE

Natural Philosophy through the Eighteenth Century & Allied Topics

CONTENTS

The Philosophical Magazine. By ALLAN FERGUSON, M.A., D.Sc., and JOHN FERGUSON, M.A., B.D.

Astronomy through the Eighteenth Century. By Sir H. SPENCER-JONES, F.R.S.

Physics in the Eighteenth Century. By Prof. HERBERT DINGLE, D.Sc.

Chemistry through the Eighteenth Century. By Prof. J. R. PARTINGTON, D.Sc.

Mathematics through the Eighteenth Century. By J. F. SCOTT, Ph.D.

Engineering and Invention in the Eighteenth Century. By Engineer-Captain EDGAR C. SMITH, O.B.E., R.N.

Scientific Instruments in the Eighteenth Century. By ROBERT S. WHIPPLE, M.I.E.E., F.Inst.P.

The Scientific Periodical from 1665 to 1798. By DOUGLAS McKIE, D.Sc., Ph.D.

Scientific Societies to the end of the Eighteenth Century. By DOUGLAS McKIE, D.Sc., Ph.D.

The Teaching of the Physical Sciences at the end of the Eighteenth Century. By F. SHERWOOD TAYLOR, Ph.D.

ALERE FLAMMAM

viii + 164 pages

15/6

POST FREE

TAYLOR & FRANCIS, LTD.

RED LION COURT, FLEET ST., LONDON, E.C.4

PRINTERS & PUBLISHERS FOR OVER 150 YEARS

XLII. *On the Fragmentations of Heavy Cosmic Ray Nuclei*

By K. GOTTSTEIN *
H. H. Wills Physical Laboratory, University of Bristol
and
Max-Planck-Institut für Physik, Göttingen †

[Received December 15, 1953]

SUMMARY

In photographic emulsions exposed at 95 000 ft. the reactions of heavy nuclei ($Z \geqslant 3$) with the nuclei of the emulsions have been studied. The charges and energies of the cosmic ray nuclei were determined both ‘ directly ’ from their multiple scattering and δ-ray densities, or from the variation of δ-ray density with range, and ‘ indirectly ’ by measuring the opening angles of the showers of fragments, and the total energy released in the reactions. A survey is given of the different types of reactions observed.

The observed frequency of occurrence of heavy fragments ($Z \geqslant 3$) produced in the encounters of heavier nuclei is in accordance with the results obtained by Peters *et al.* The charge distribution at the top of the atmosphere is calculated from the one measured below material (air, glass, emulsion) corresponding to 15–40 g/cm^2 of air, using the collision cross section as found by Peters. The influence of energy loss by ionization is considered. The resulting charge distribution is in approximate agreement with the one measured by Dainton, Fowler and Kent, the nuclei Li, Be, B being about as frequent as C, N, O, F. The energy distribution agrees with the one obtained by Dainton, Fowler and Kent.

The frequency of meson production in collisions of heavy nuclei is studied.

§1. INTRODUCTION

THE investigations on the charge spectrum of the primary cosmic radiation made by different authors during the past few years by the photographic method have led to results not fully in agreement with each other. Thus Bradt and Peters (1950) and Peters (1952) came to the conclusion that the flux of Li, Be and B nuclei at a geomagnetic latitude of 30° is 10% or less of the flux of C and O nuclei, and that the Li, Be and B nuclei observed under 15–20 g/cm^2 of air are mostly fragments of heavier particles having suffered collisions inside the atmosphere. Measurements under latitudes of 42° and 55° confirmed this result (Kaplon *et al.* 1952). According to the observations of Dainton, Fowler and Kent (1951, 1952), however, the flux of the Be and B nuclei at the top of the atmosphere is about 83% of the flux of C, N, O and F. The proportion of the Li, Be, B group among the primary nuclei is of considerable importance for the conceptions on the origin of the cosmic radiation (Bradt and Peters 1950, Dainton *et al.* 1952, Biermann 1953).

---

* Now at the Max-Planck-Institut für Physik, Göttingen, Germany.
† Communicated by Professor C. F. Powell, F.R.S.

Peters and collaborators determined the charges and energies of particles by measuring the grain or δ-ray densities and the residual ranges in stacks consisting of alternating layers of nuclear plates and brass or, for high energies, the opening and relative scattering angles between the tracks of α-particle fragments originating inside the brass layers in collisions of their ' mother nuclei '. Disadvantages of this method are the inaccessibility to direct observation of events occurring within the brass, and, for higher energies, the limitation imposed by the fact that at least 3 α-particles must occur as fragments in collisions of primary nuclei for the method to be applicable. The Bristol workers obtained their results by measuring the grain and δ-ray densities and the mean angles of multiple scattering. In consequence they were restricted to tracks of a length of at least 4 mm per emulsion.

The method adopted in the present investigation has already been briefly described (Gottstein 1953 a). It consists in the systematic analysis of the collisions of heavy nuclei ($Z \geqslant 3$) occurring in the emulsion layers of nuclear plates. Even if the tracks of heavy nuclei are too short for scattering measurements to be feasible, they are very frequently long enough to have a number of δ-rays high enough for their density to be measured with not too large a statistical error. Considering the numbers, masses and energies of the fragments, both of the incoming and the target nucleus, and allowing for neutral particles, one obtains the charge and energy of the primary in most cases with good accuracy. A further advantage of the method is the direct visibility of the events concerned thus permitting a detailed study to be made of the phenomena connected with the disintegrations. A disadvantage is its restriction to nuclei suffering collisions.

§2. EXPERIMENTAL PROCEDURE

The emulsions used for this investigation (Ilford G5, 400 μ thick) are part of those in which Dainton, Fowler and Kent have carried out their measurements. They had been exposed in a vertical position at 95 000 ft. at a geomagnetic latitude of 55°. All ' stars ' containing ' heavy ' tracks with δ-ray densities of at least 1 δ-ray per 100 microns have been considered, provided these tracks lay in the upper hemisphere and were obviously not those of slow secondary particles. Since this δ-ray density corresponds to the minimum value for Li, *all* stars being connected with nuclei heavier than He have thus been included. A ' star ' in this connection was defined as an event in which at least one further track (not being a δ-ray) branched off from the heavy track. 118 such events were found, the number of tracks per ' star ', apart from the heavy primary, ranging from 2 to 75. In all there were 2088 tracks in the events.

In measuring the δ-ray densities the convention of Dainton *et al.* was adopted, counting all δ-rays with 4 or more grains.

Grain densities, which were measured for all the 1322 ‘ thin ’ and ‘ grey ’ tracks connected with the events were normalized with regard to the minimum grain densities in the respective regions of the emulsions, as obtained from the tracks of energetic electron–positron pairs.

The mean angle of multiple scattering was measured on all tracks of heavy primaries and their fragments as well as on all ‘ thin ’ tracks belonging to the events whenever the lengths of the tracks allowed this. The coordinate method of Fowler was used (Fowler 1950, Gottstein *et al.* 1951) and the influences of spurious scattering as caused by the distribution of silver grains, mechanical imperfections of the microscope stage, distortion of the emulsion and subjective errors were as far as possible eliminated by the methods previously described (Menon *et al.* 1951, Gottstein 1953 b).

In principle, there exist several methods for the identification of heavy nuclei suffering collisions, and as many of them as possible have been applied in individual cases. In general, there was good agreement between the results rendered by different methods.

The ‘ direct ’ way consists in the measurement of δ-ray density and multiple scattering (Dainton *et al.* 1951, 1952), or of the variation of δ-ray density with the residual range in the stacks of plates (see fig. 4 from Dainton *et al.* 1952). ‘ Indirectly ’, one can obtain the charge and energy of the primary nucleus solely by measuring the δ-ray density of its track, the total energy of the particle being derived from an analysis of the disintegration process. This can be done in one, or several, of the following ways :

(a) *Measurement of the Angles θ between the Tracks of the Fragments and the Track of the Primary Particle*

Assuming the mean kinetic energy of protons and α-particles in the centre of mass system to be 12 MeV and the greatest energy occurring with noticeable intensity to be 30 MeV (Harding *et al.* 1949) one obtains for protons

$$\sqrt{(\overline{\theta^2})}=\frac{0\cdot 12}{\epsilon_0},$$

$$tg\,\theta_{\max}=\frac{0\cdot 25}{\epsilon_0},$$

and for α-particles (Peters 1952)

$$\sqrt{(\overline{\theta^2})}=\frac{0\cdot 06}{\epsilon_0},$$

$$tg\,\theta_{\max}=\frac{0\cdot 12}{\epsilon_0},$$

$\theta_{\max}$ denoting the greatest angle possible in the laboratory system, and ϵ_0 the kinetic energy per nucleon of the heavy primary. To obtain ϵ_0 in BeV, θ is to be measured in radians.

Because of the great statistical fluctuations in the case of small numbers of fragments this method frequently only gives an upper limit for ϵ_0.

(b) *Determination of the Velocities of Fragments*

If there is a narrow ' jet ' of particles continuing in the direction of the primary, obviously representing the fragments (as e.g., in Plate 8), one can in many cases assume that their velocity equals that of the primary. If the jet contains particles with grain density between one and four times minimum, grain counting alone gives the velocity. In those cases where the fragments are in the minimum ionization region without scattering measurements being possible, grain counting gives only a lower limit for the velocity, but this is sufficient for the identification of the nucleus (fig. 4 from Dainton *et al.* 1952).

(c) *The Smallest Grain Density*

If there is no narrow ' jet ' of fragments, but there are one or more ' thin ' or ' grey ' tracks in the event, then the assumption may be used that the velocity of the primary was at least as high as that of the fastest of the secondary particles (i.e., that with the smallest grain density). In general, the fixing of this lower limit is sufficient for the identification of the primary nucleus. The correctness of the above assumption was checked in several cases where the primary velocity could be determined independently.

(d) *Energy Balance*

The total energy appearing in the reaction must equal the primary energy. From fig. 4 given by Dainton *et al.* (1952) one determines which nucleus having the δ-ray density measured, could have supplied the required energy. In many cases the result is unambiguous.

In calculating the total energy of the reaction, a mean energy of 20 MEV (including binding energy) was assigned to every ' black ' evaporation track. The mean energy of a proportion of the ' grey ' tracks was found to be 150 MEV (per nucleon) and this value was then used for all ' grey ' tracks. ' Thin ' tracks, whose scattering could not be measured, were assigned the energy corresponding to a proton of their grain density. ' Minimum ' tracks were assumed to have at least an energy of 1 to 1·5 BEV.

To take into account neutral particles, the total energy was assumed to equal 2·2 times the sum of the energies of all charged particles.

In some cases ' partial balances ' were set up, only part of the fragments of the primary nucleus being recognizable as such. Apparently one part of the fragments had spent their energies for the disintegration of the target nucleus, the remaining fragments having lost relatively little energy. It was investigated in these cases which assumptions on the nature of the individual particles could explain the measurements.

§3. RESULTS

A. *General Survey*

As mentioned by Bradt and Peters (1949), three kinds of collisions of heavy nuclei may be distinguished phenomenologically :

(*a*) The heavy nucleus, having split up the target nucleus, continues without loss of charge.

(*b*) Several protons and α-particles are split off the nucleus in the collision but there is still a heavy remnant ($Z \geqslant 3$) continuing approximately in the original direction.

(*c*) The heavy nucleus is completely split up into protons and α-particles, or even merely into protons.

Plates 7–9 show examples of disintegrations of type (*b*) and (*c*).*

Of the 118 events on which this investigation is based, 2 belonged to type (*a*), 31 to type (*b*) and 75 to type (*c*). In 7 cases the primary proved to be a relatively slow α-particle, in 2 cases the experimental conditions did not allow the determination of the charge of the primary nucleus, and in one case the particle having caused the ' δ-ray track ' was found to be a fragment of the target nucleus.

In table 1 are compiled for each primary charge number the fragments found, as far as they could be identified. Each compartment corresponds to one collision and contains the numbers and types (chemical symbols) of fragments identified. The letter ' H ' in the lower left corner of the compartments indicates that there were more than 8 ' black ' or ' grey ' tracks in the events apart from those caused by the fragments, so that the target nucleus must have been a ' heavy ' one (S, Br, Ag, J). The letter ' L ' denotes events with 8 or less ' heavy ' tracks, i.e. collisions with H, C, N, O or ' glancing ' collisions with heavier nuclei. The figure in the right lower corner gives the energies per nucleon of the primaries in MEV. The statistical error in the determination of charges $Z \geqslant 3$ was smaller than 1 in more than 50% and smaller than 2 in more than 80% of the cases. (5 of the 9 nuclei called F could possibly be O or Ne.) In those cases where the compartments contain a dash, the fragments of the primary could not be distinguished from those of the target nucleus. For the lighter nuclei almost all possible modes of disintegration are realized, as table 1 shows. There is no significant preponderance of particular ways of splitting for any type of nucleus. In view of the variety of target nuclei, impact parameters and energies, this is not surprising.

Of 414 elementary charges carried by the primaries to ' H ' events only 309 reappear among the fragments, whereas for ' L ' events the corresponding figures are 427 out of 447. (The incoming particles are more strongly affected by collisions with heavy target nuclei than by collisions with light ones.)

---

* The events shown in Plates 7–9 were found in emulsions exposed in a balloon flight carried out at Göttingen. Because of the differences in latitude and altitude reached they are not included in table 1.

Table 1

Li	Be	B	C	N	O	F	Ne	Na	Mg	Al	Si	P	S	Cl	A	Ca	V
3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	20	23
2H L 70	1Li L 110	2He, 1H L 400	2He, 2H L 50	3He, 1H L 45	2Li, 2H L 250	1N, 1He L 40	1O, 1He L 400	1N, 4H L ≥ 5000	3He, 6H L 350	2He, 9H L 2000	1F, 2He, 1H L 700	4He, 7H L ≥ 2000	1Al, 1He, 1H H ≥ 2000	2He, 9H H 800	1S, 2H L 300	1O, 4He, 4H L 6500	1Cl, 1He, 4H L 250
— L 100	1Li, 1H L 250	1He, 3H L 450	2He, 2H L 200	3He, 1H L 400	1C, 1He L 800	4He, 1H L 140	1O, 1He L 600		3He, 6H L 1500	6H H 200	2He, 8H H ≥ 1000	1N, 2He, 1H H 320				2B, 1Be, 1Li H 800	
1He, 1H L >250	2He L 300	1Li, 1He L 500	2He, 2H L 600	1Be, 1He, 1H L 500	3He, 2H L >1500	9H L >1500	4He, 2H L 800		1He, 10H H 600	2He, 9H H 3000	1Li, 1He, 9H H >1500						
1He, 1H L 500	2H L 300	2He, 1H L ≥800	2He, 2H L 600	2He, 3H L 700	1B, 3H H 300	— H 170	1C, 2He L 1000		3He, 6H H ≥1500		4He, 6H H 2000						
1H H 300	2He L 1000	2He, 1H L 1000	3He L 700	1He, 5H L 2000	2Li* H 500	1Be, 1Li H 250	1Be, 6H L 1000		1He, 10H H ≥1500								
	1He, 2H L >2000	1Li, 2H L ≥1500	2He, 2H L 800	2He, 3H L 10000	— H 600	1He, 7H H 700	2He, 6H L ≥1500		2Be, 2He H >2000								
	2He L 4000	3H H 300	1Be, 1He L 900	— H 250		7H H ≥ 1000	2He, 6H L ≥2000										
	4H L 6000	2He, 1H H 600	1Be, 2H L 1000	— H 350		9H H 2500	1C, 1He H 450										
	4H H 300	2He, 1H H 600	— L > 1000	1He, 5H H ≥ 800		1He, 7H H ≥3500	3He, 3H H ≥1500										
	1He, 2H H 1500	2H H 900	1Be, 1He L ≥ 1000	5H H 900													
	4H H 4000	— H 1500	2He, 2H L 1800														
		1He, 3H H 1500	1Li, 1He, 1H L 2500														
		5H H ≥ 4000	— H 70														
			1He, 2H H 200														
			— H 300														
			— H 350														
			— H 400														
			— H 1500														
			6H H >1500														
			1B, 1H H 2000														
			1He, 4H H >2000														

Table 2 shows the numbers of different types of identified fragments for primary kinetic energies smaller than and equal to or greater than 1 Bev per nucleon, separately for H and L events.

Table 2

	Total No. of nuclei	Total No. of identified fragments				
		H	He	Li, Be, B	CNOF	$Z \geqslant 10$
			$E < 1$ Bev per nucleon			
H events	27	62	13	9	2	0
L events	34	39	51	7	5	2
			$E \geqslant 1$ Bev per nucleon			
H events	21	100	22	4	0	1
L events	24	81	36	5	3	0

As can be seen, the primary nuclei are more thoroughly split up (relatively less α-particles and more protons appearing) in collisions with heavy target nuclei (H-events) than in those with light ones. This also remains true if one considers separately the three groups of primary nuclei with $3 \leqslant Z \leqslant 5$, $6 \leqslant Z \leqslant 9$ and $Z \geqslant 10$. (Following the terminology of Bradt and Peters we shall call the nuclei belonging to these groups A-, B-, and C-nuclei, respectively.) The effect is less strongly marked for high energies where even collisions with lighter target nuclei result in a more complete break-up of the incoming particles.

One notices further that the relative frequency of occurrence of α-particles as fragments is, at least for the L events, dependent on energy.

Table 3 shows the frequency ratios of α-particles to singly charged particles.

Table 3

$$\frac{\text{Number of } \alpha\text{-particles}}{\text{Number of singly charged particles}}$$

	A-nuclei	B-nuclei	C-nuclei
H events	0·21	0·09	0·31
L events	0·81	1·08	0·49

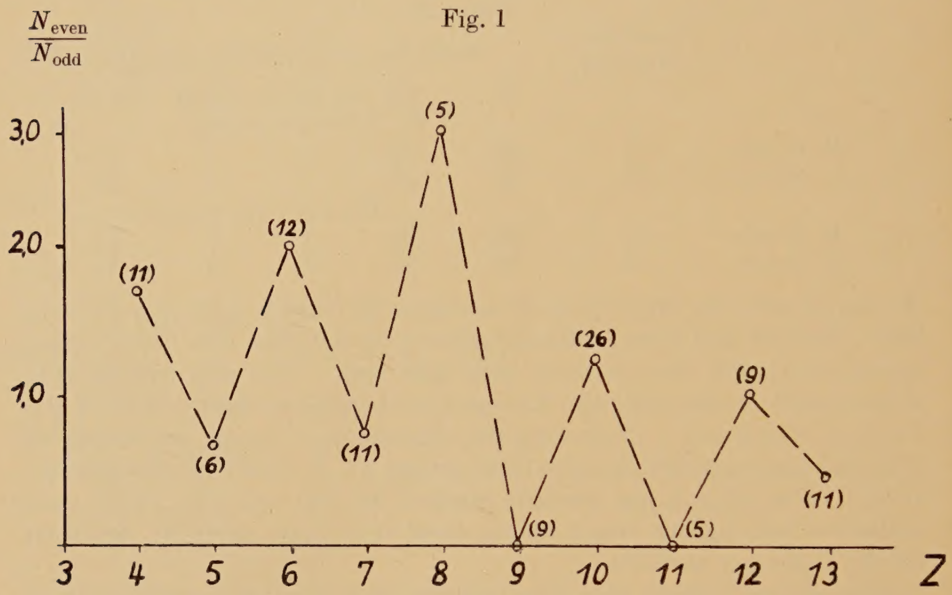
These values are in good agreement with those obtained by Perkins (1949, 1950 a) in the analysis of the evaporation products of nuclei stationary in the emulsion, i.e. ~1 for light nuclei (C, N, O) and 0·4–0·6 for heavy nuclei, the corresponding ratios in table 3 being 1·08 and 0·49.*

Figure 1 shows the ratio of elementary charges appearing in ‘ even ’ fragments (He, Be, C, . . .) to that in ‘ odd ’ fragments (H, Li, B, . . .) as a function of primary charge for L events and kinetic energies $\geqslant 1$ Bev per nucleon, these events corresponding roughly, in the opposite frame of reference, to the ‘ stars ’ with energetic ‘ light ’ primaries. Although the statistical fluctuations to be expected are rather large, there is a distinct tendency for the probability of emission of 'even' fragments to be higher for 'even' nuclei than for 'odd' ones. A similar regularity is neither recognizable for low energies, nor for H events in general.

---

* Since the collision partners of the nuclei in the reactions investigated by Perkins are single nucleons, our L events should be compared with those reactions, the light and heavy nuclei of Perkins corresponding to our B and C nuclei.

Fig. 1



Number of fragments with even charge number divided by the number of fragments with odd charge number as a function of charge number of 'mother nucleus'. (L-events, kinetic energy per nucleon ⩾ 1 Bev.) The figures in brackets denote the numbers of charged fragments on which the individual experimental points are based.

B. *The Charge Distribution of the Heavy Nuclei at the Top of the Atmosphere*

The number of A-, B-, and C-nuclei in a parallel beam having traversed h g/cm² of matter is given by the formulae

$$\begin{aligned}
N_A(h)=\;&N_A(0)\exp(-h/\lambda_A)+N_B(0)\,p^A_B\,\frac{\lambda_A\Lambda_B}{\lambda_A-\lambda_B}\\
&\times[\exp(-h/\lambda_A)-\exp(-h/\lambda_B)]+N_C(0)\,p^A_C\,\frac{\lambda_A\Lambda_C}{\lambda_A-\lambda_C}\\
&\times[\exp(-h/\lambda_A)-\exp(-h/\lambda_C)]\\
&+\left\{N_C(0)\,p^B_C\,p^A_B\,\frac{\lambda_A\Lambda_B}{\lambda_A-\lambda_B}\left[\frac{\lambda_A\Lambda_C}{\lambda_A-\lambda_C}[\exp(-h/\lambda_A)-\exp(-h/\lambda_C)]\right.\right.\\
&\left.\left.-\frac{\lambda_B\Lambda_C}{\lambda_B-\lambda_C}[\exp(-h/\lambda_B)-\exp(-h/\lambda_C)]\right]\right\}, \qquad (1)
\end{aligned}$$

$$\begin{aligned}
N_B(h)=\;&N_B(0)\exp(-h/\lambda_B)+N_C(0)\,p^B_C\,\frac{\lambda_B\Lambda_C}{\lambda_B-\lambda_C}\\
&\times[\exp(-h/\lambda_B)-\exp(-h/\lambda_C)], \qquad (2)
\end{aligned}$$

$$N_C(h)=N_C(0)\exp(-h/\lambda_C). \qquad (3)$$

$N_{\mathrm{A}}(0)$, $N_{\mathrm{B}}(0)$, $N_{\mathrm{C}}(0)$ denote the number of A-, B-, and C-nuclei at the top of the atmosphere, λ_{A}, λ_{B}, λ_{C} the ‘ absorption mean free paths ’ for the different groups of nuclei ; $p_{\underset{\mathrm{k}}{\mathrm{i}}}$ is the probability for the occurrence of an i-type fragment in the break-up of a k-nucleus.

$\Lambda_{\mathrm{B,\,C}}$ is the ratio of the mean free paths for absorption and collision, for B- and C-nuclei

$$\Lambda_{\mathrm{i}}=\frac{1}{1-p_{\underset{\mathrm{i}}{\mathrm{i}}}}\,.$$

The quantities $p_{\underset{\mathrm{k}}{\mathrm{i}}}$ may be derived empirically from table 1.

It follows, for all events :

$$\begin{array}{ll}
p_{\underset{\mathrm{B}}{\mathrm{A}}}=0{\cdot}28 & \qquad p_{\underset{\mathrm{Total}}{\mathrm{A}}}=0{\cdot}24\\
p_{\underset{\mathrm{C}}{\mathrm{A}}}=0{\cdot}26 & \qquad p_{\underset{\mathrm{B+C}}{\mathrm{B}}}=0{\cdot}13
\end{array}$$

For L events separately :

$$\begin{array}{ll}
p_{\underset{\mathrm{B}}{\mathrm{A}}}=0{\cdot}29 & \qquad p_{\underset{\mathrm{Total}}{\mathrm{A}}}=0{\cdot}22\\
p_{\underset{\mathrm{C}}{\mathrm{A}}}=0{\cdot}06 & \qquad p_{\underset{\mathrm{B+C}}{\mathrm{B}}}=0{\cdot}20
\end{array}$$

Within the limits of statistical errors these values are in agreement with those observed by Bradt and Peters (1950) for collisions in glass ($p_{\underset{\mathrm{B}}{\mathrm{A}}}=0{\cdot}23$; $p_{\underset{\mathrm{C}}{\mathrm{A}}}=0{\cdot}24$; $p_{\underset{\mathrm{B+C}}{\mathrm{A}}}=0{\cdot}23$; $p_{\underset{\mathrm{B+C}}{(\mathrm{CNO})}}=0{\cdot}22$) and by Perkins (1950 b) who found that in C2 emulsions the average number of heavy fragments ($Z\geqslant 3$) emitted from stars with 7–11 prongs is 0·20.

Since the formulae (1), (2) and (3) are valid for a parallel beam, the thicknesses of material traversed by each individual particle in air, glass and emulsion up to its disintegration have been determined. The mean thickness of material traversed by the particles corresponds to 25·7 g/cm^2 of air, if one transforms the individual values of h into those valid for a homogeneous medium of air, using the cross sections for the different materials as given by Peters (1952).

In order to obtain the true frequency ratios of the different sorts of nuclei, one has to divide their absolute numbers, as found experimentally, by their cross sections for collisions. Setting the correction factor for the frequency of occurrence of B-nuclei equal to 1, one obtains that the number of A-nuclei found should be multiplied by 1·2 and that of the C-nuclei by 0·75.

The ‘ projected ’ lengths inside the emulsion layers where the fragmentations occurred have been measured for all primary tracks included in this investigation. Table 4 shows the numbers of tracks having different lengths. Since the angular distributions of the different types of tracks may be assumed to be identical the same should apply to their length distributions. It thus follows from table 4 that the relative numbers of short tracks which have been overlooked in the scanning of the emulsions are considerably greater for A- than for B- or C-nuclei. We have therefore used the assumption that only the values in the last column of table 4 are unaffected by this systematic error.

Table 4

Projected track lengths (μ)	<500	500–3000	>3000
A-Nuclei	2	13	14
B-Nuclei	9	25	12
C-Nuclei	11	14	6

Using for λ and Λ the values calculated from the geometrical cross sections and the $p_{\substack{i\\k}}$ as given by Bradt and Peters (1950) one obtains ($\bar{h}=25\cdot 7$ g/cm^2) an abundance ratio of Beryllium and Boron nuclei to ‘B-nuclei’ equal to 0·79. This is in good agreement with the results of Dainton *et al.* (0·83) whereas according to Peters* the incoming flux of A-nuclei (including Li) may be assumed to be smaller than 0·1 times that of B-nuclei.

One obtains further $N_B(0)/N_C(0)=1\cdot 7\pm 0\cdot 5$, a value somewhat smaller than that found by the other workers (3·0).

C. *The Energy Distribution of the Heavy Nuclei at the Top of the Atmosphere*

Having determined the thickness of material traversed by each individual nucleus before its disintegration, its energy at the top of the atmosphere was calculated from the range–energy relation.† Table 5 shows the resulting numbers of nuclei in different energy intervals at the top of the atmosphere.

Table 5

Energy per nucleon (MEV per nucleon)	<300	300–400	400–600	600–800	800–1000	1000–3000	>3000
A-Nuclei	0	4	6	5	2	7	4
B-Nuclei	0	5	11	5	6	17	2
C-Nuclei	—	—	1	5	5	17	3

Particles with energies below 300 MEV/nucleon do not occur among the A- and B-nuclei; the ‘geomagnetic cut-off energy’ at the latitude of exposure (55°N) is about 350 MEV/nucleon (for $M=2Z$).

The values of table 5 do not yet represent the true energy distribution at the top of the atmosphere because some of the incident nuclei will have lost their entire energy by ionization before reaching the stack of plates. To take into account this effect, the distribution of thicknesses of material traversed by particles with energies $\geqslant 1$ BEV/nucleon has been determined. Nuclei with these high energies will not be stopped by ionization loss even when having to traverse the greatest thicknesses occurring in our experimental arrangement. It was then assumed that the distributions of thicknesses traversed by particles of all energies which would have arrived at the stack of plates but for ionization loss, were identical. Table 6 shows the percentages thus obtained of nuclei of different energies (at the top of the atmosphere) which were unable to reach the emulsions because of ionization loss.

---

* As Professor Peters kindly pointed out to us, the value of N_A/N_B observed by him and his collaborators under 15–20 g/cm^2 of air was 0·275 and not 0·112, as was erroneously assumed by us in a previous communication (Gottstein 1953 a).

† The fact was neglected that some of the nuclei must have been fragments of heavier nuclei from collisions inside the atmosphere, but since their number—according to §3 B—is not more than 20% of the total, the error involved is not serious.

Table 6

Energy per nucleon (MEV per nucleon)	300–400	400–600	600–800	800–1000	1000–3000	>3000
A-Nuclei	11	0	0	0	0	0
B-Nuclei	52	12	0	0	0	0
C-Nuclei	100	60	17	4	0	0

If one applies a corresponding correction to table 5 it follows that 9% of all nuclei with $Z \geqslant 3$ have kinetic energies >3000 MEV/nucleon, and 42% kinetic energies >1000 MEV/nucleon. Since ‘ spurious scattering ’ of the tracks makes it difficult in some cases to measure accurately scattering angles smaller than 0·01 degrees per 100 microns and opening angles of fragment showers smaller than 1 degree these proportions may be taken to represent lower limits. In table 7 the results are compared with those of previous workers, for equal latitudes.

Table 7

Authors	Proportions of heavy nuclei at the top of the atmosphere with kinetic energies	
	>1000 MEV/nucleon	>3000 MEV/nucleon
Peters	55%	22%
Dainton, Fowler, Kent	45%	8%
Gottstein	42%	9%

D. *Meson Production in the Collisions of Heavy Nuclei*

The direct determination by multiple scattering and grain-density measurements of the masses of singly charged particles is only possible if their ionization has not yet reached the ‘ plateau ’ value and if their tracks within the emulsions are of sufficient lengths. In this manner a number of π mesons could be identified among the particles arising from the collisions of heavy nuclei. In a few cases particles have been observed with mass values similar to those reported by Daniel *et al.* (1952) for the κ-mesons, but the magnitude of the statistical errors does not allow definite conclusions to be drawn.

Whenever mass determinations were not possible, the proportion of mesons among the ‘ shower ’ particles was estimated in the following way :

It was assumed that the number of protons among them is given by the difference between the number of charges on the incoming nucleus and the total charge of the more than singly-charged fragments, the latter being easily identifiable. The remaining ‘ thin ’ tracks were regarded as mesons. If the number of the ‘ shower ’ particles was equal to, or smaller than the number of primary charges, it was assumed that no mesons had been produced.

Of 20 nuclei with energies between 600 and 1000 Mev/nucleon, 6 produce mesons, 5 of the 6 events being of the ‘ L ’ type. The average number of charged mesons in this energy range is 15/20=0·75.

Table 8. (Primary energies $\geqslant 1$ Bev/nucleon)

Charge	H events Number of Events	H events Number of Mesons	L events Number of Events	L events Number of Mesons
3, 4	2	9	4	14
5, 6	7	19	7	13
7, 8			3	10
9, 10	4	19	5	47
11, 12	3	2	2	34
13, 14	4	41	1	11
15, 16	1	7	1	0
19, 20			1	4

For energies $\geqslant 1$ Bev/nucleon 33 out of 45 nuclei produce mesons. In 10 out of the remaining 12 cases the primary charge equals the total charge on the singly and multiply charged ‘ relativistic ’ particles. The mean number of charged mesons per event for these energies is

for 21 H events : 4·6

for 24 L events : 5·5

There is no marked difference in the figures for H and L events, in agreement with the predictions of the theory of multiple meson production (Heisenberg 1936, 1952). This is borne out by Table 8.

ACKNOWLEDGMENTS

The author wishes to express his gratitude to Professor C. F. Powell, F.R.S., for the hospitality extended to him in the H. H. Wills Physical Laboratory in which this investigation was begun, and to Mr. P. H. Fowler for valuable discussions and suggestions. His thanks are also due to Dr. B. M. Anand for his assistance with some of the scattering measurements and δ-ray counts, and to the Bristol team of observers who scanned the plates. The microphotographs were made by Mrs. C. Zöllner (Göttingen). Drs. M. Ceccarelli and N. Barford have kindly read the manuscript.

It is a pleasure to thank Professors W. Heisenberg and K. Wirtz for facilities for continuing this work at the Max-Planck-Institut für Physik, Göttingen.

REFERENCES

BIERMANN, L., 1953, *Annual Review Nuclear Science*, Vol. 2.

BRADT, H. L., and PETERS, B., 1949, *Phys. Rev.*, **75,** 1779 ; 1950, *Ibid.*, **80,** 943.

DAINTON, A. D., FOWLER, P. H., and KENT, D. W., 1951, *Phil. Mag.*, **42,** 317 ; 1952, *Ibid.*, **43,** 729.

DANIEL, R., DAVIES, J. H., MULVEY, J. H., and PERKINS, D. H., 1952, *Phil. Mag.*, **43,** 753.

FOWLER, P. H., 1950, *Phil. Mag.*, **41,** 169.

GOTTSTEIN, K., 1953 a, *Naturwiss.*, **40,** 104 ; 1953 b, Appendix 3 to *Kosmische Strahlung* by W. Heisenberg, 2nd. ed. (Berlin : Springer-Verlag 1953).

GOTTSTEIN, K., MENON, M. G. K., MULVEY, J. H., O'CEALLAIGH, C., and ROCHAT, O., 1951, *Phil. Mag.*, **42,** 708.

HARDING, J. B., LATTIMORE, S., and PERKINS, D. H., 1949, *Proc. Roy. Soc.* A, **196,** 325.

HEISENBERG, W., 1936, *Zs. f. Phys.*, **101,** 533 ; 1952, *Ibid.*, **133,** 65.

KAPLON, M. F., PETERS, B., REYNOLDS, H. L., and RITSON, D. M., 1952, *Phys. Rev.*, **85,** 295.

MENON, M. G. K., O'CEALLAIGH, C., and ROCHAT, O., 1951, *Phil. Mag.*, **42,** 932.

PERKINS, D. H., 1949, *Phil. Mag.*, **40,** 601 ; 1950 a, *Ibid.*, **41,** 138 ; 1950 b, *Proc. Roy. Soc.* A, **203,** 399.

PETERS, B., 1952, *Progress in Cosmic Ray Physics* (Amsterdam : North Holland Publishing Company).

XLIII. *The Ideal Crystal at Absolute Zero*

By L. Salter*
The Clarendon Laboratory, Oxford†

[Received December 28, 1953]

Abstract

Theoretical expressions for the internal energy, equilibrium volume, and elastic constants of the rare gas solids at absolute zero are derived, using a recently proposed approximation for the zero point energy of a crystal. The calculations show that, on the basis of a central force model, the zero point energy destroys the equality $c_{12}=c_{44}$ (Cauchy relation) for these solids.

§ 1

Recently a new expression for the zero point energy of crystals has been proposed (Domb and Salter 1952):

$$U_z=(9/8)Nk\Theta_\infty \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (1)$$

where Θ_∞, the limiting value of the equivalent Debye Θ as $T\rightarrow\infty$, replaces Θ_0, the limiting value as $T\rightarrow 0$. It was there shown that Θ_∞ is very simply related to the trace of the dynamical matrix for the lattice vibration problem, and that for the Bravais cubic lattices in the case of nearest and next nearest neighbour interactions eqn. (1) gives quite good agreement with the more exact value calculated by the use of the moments of the frequency spectrum.

Assuming, then, that these results may hold more generally (that is, for all neighbour interactions and for other lattice structures besides the three cubic types), it is of interest to investigate what changes this new zero point energy expression implies in the calculations of quantities of interest—internal energy, equilibrium volume, elastic constants—at the absolute zero of temperature.

As an example of such calculations we note the work of de Boer and Blaisse (1948) with regard to the inert gas solids; they attempted to correlate theoretical values with experiment by the addition of a zero point energy term U_z to the usual lattice energy term U_0. Their expression for U_z involves the elastic constants (c_{11}, c_{12}, and c_{44} for the cubic crystals) in a rather complicated fashion, so that numerical computations are heavy work. There is the further disadvantage that the expressions for the elastic constants used are the usual ones (Voigt 1928, Born 1940) computed from the static lattice energy term U_0 only, instead of from the free energy U_0+U_z. In the following we derive, by a unified approach based on the use of the correct free energy, expressions for the quantities of interest at absolute zero, including the elastic constants themselves. The resulting equations, simpler in form through the use of Θ_∞, confirm and extend the results of de Boer and Blaisse. In addition an interesting result is obtained with respect to the oft-discussed question of the fulfilment of the Cauchy relations (Stakgold 1950).

---

* Communicated by Professor F. Simon, F.R.S.

† Now at Wabash College, Crawfordsville, Indiana, U.S.A.

We use the formalism of Born (Born 1940) to develop the free energy of the crystal at $T=0$ with respect to small homogeneous deformations of the lattice and thus obtain, as the coefficients of the strain components, the equation of state and the elastic constants. Central forces are assumed from the beginning and this allows expression of the results in terms of reduced units (de Boer and Blaisse 1948).

§ 2

We consider the case of the three Bravais cubic lattices. The lattice points of the undeformed lattice are given by $\mathbf{r}^{0l}=(l_1a,\ l_2a,\ l_3a)$; under a deformation distorting the cube of side a into a parallelepiped the lattice points of the distorted lattice are given by $\mathbf{r}^l=l_1\mathbf{a}_1+l_2\mathbf{a}_2+l_3\mathbf{a}_3$. Defining ρ^l by the equation $|\mathbf{r}^l|^2=|\mathbf{r}^{0l}|^2+2\rho^l$, we can write for ρ^l, $\rho^l=(a^2/2)\sum\limits_{i=1}^{3}\sum\limits_{j=1}^{3} l_il_je_{ij}$, the e_{ij} being the components of the strain tensor in Brillouin's notation. In terms of the $\mathbf{a}_i$, $e_{ij}=(\mathbf{a}_i\,.\,\mathbf{a}_j-a^2\delta_{ij})/a^2$.

Considering for the moment the unstrained lattice, the equations of motion are

$$m\ddot{u}_\alpha{}^l=\sum_{l'=1}^{N}\sum_{\beta=1}^{3}\phi_{\alpha\beta}^{l-l'}u_\beta{}^{l'} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (2)$$

where $\phi^l_{\alpha\beta}=\delta_{\alpha\beta}D\phi(|\mathbf{r}^{0l}|)+D^2\phi(|\mathbf{r}^{0l}|)$, $D\equiv\frac{1}{r}\frac{\partial}{\partial r}$, $\phi^0_{\alpha\beta}=-\sum\limits_{l>0}\phi^l_{\alpha\beta}$.

We can assume solutions of the type

$$u_\alpha{}^l=U_\alpha{}^l\exp(-i\omega t), \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (3)$$

and also the Born–von Kármán plane wave type

$$\mu_\alpha{}^l=V_\alpha\exp[i(l\alpha)]\,.\,\exp(-i\omega t),\ (l\alpha)=l_1\alpha_1+l_2\alpha_2+l_3\alpha_3. \quad . \quad . \quad (4)$$

Θ_∞ is given by the expression (Domb and Salter 1952)

$$\Theta_\infty=(h/k)[(5/3)\mu_2]^{1/2},\ \mu_2=(3N)^{-1}Tr[A] \quad . \quad . \quad . \quad . \quad (5)$$

where μ_2 is the second moment of the frequency distribution and $Tr[A]$ is the trace of the dynamical matrix occurring in the secular equation for the frequencies, $|A-U^2I|=0$. The first solution leads directly to the result

$$Tr[A]=\frac{N}{4\pi^2m}\sum_\alpha\sum_{l>0}\phi^l_{\alpha\alpha} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (6)$$

and the second moment of the frequency distribution is thus

$$\mu_2=(12\pi^2m)^{-1}\sum_{l>0}\left(\frac{\partial^2}{\partial r^2}+\frac{2}{r}\frac{\partial}{\partial r}\right)\phi(|\mathbf{r}^{0l}|). \quad . \quad . \quad . \quad (7)$$

We get, of course, the same result by constructing the 3×3 matrix $[B]$ for the plane wave solution; in this case $\mu_2=\langle Tr[B]\rangle/3$, $\langle Tr[B]\rangle$ being the value of the trace averaged over all allowable values of the wave phases α_i.

The free energy at absolute zero of the unstrained lattice will consist of the static lattice energy plus the zero point energy term $U_z=(9/8)Nk\Theta_\infty$ and by the above relations

$$U_z=(3/16\pi)Nk\left[\frac{5}{m}\sum_{l>0}|\nabla^2\phi(r)|_{|\mathbf{r}^{0l}|}\right]^{1/2} \quad . \quad . \quad . \quad . \quad (8)$$

and
$$F/N=\tfrac{1}{2}\sum_{l>0}\phi(r^0)+\frac{3k}{16\pi}\left[\frac{5}{m}\sum_{l>0}\nabla^2\phi(r^l)\right]^{1/2},\ r^l=|\mathbf{r}^{0l}|. \quad . \quad . \quad (9)$$

We seek an expansion of the free energy per molecule in terms of the strain components; using the expansion (valid for any function $f(|\mathbf{r}^{0l}|)$ of the lattice sites)

$$f\{\sqrt{(r^2+2\rho)}\}=f(r)+\rho Df(r)+(\rho^2/2\,!)D^2f(r)+\ldots., \quad . \quad . \quad (10)$$

we can easily expand both the quantities $\phi(r^l)$ and $(\phi''+2/r\,\phi')r_l$ about the undistorted lattice sites $\mathbf{r}^{0l}$. Expanding, in turn, the radical up to terms of second order in ρ^l, using the result $\rho^l=(a^2/2)\sum_{ij}l_il_je_{ij}$, taking into account cubic symmetry, and collecting terms in the e_{ij}, we obtain

$$\begin{aligned}\frac{F}{N}=&\tfrac{1}{2}\sum_{l>0}\phi(r^{0l})+\frac{3\hbar}{8}\left(\frac{5S}{m}\right)^{1/2}\\&+\frac{a^2}{4}\left\{S_1^{(0)}+\frac{3\hbar}{8}\left(\frac{5}{mS}\right)^{1/2}\sum_{l>0}l_1^2Dg(r^{0l})\right\}(e_{11}+e_{22}+e_{33})\\&+\frac{a^4}{16}\left\{S_{11}^{(0)}+\frac{3\hbar}{8}\left(\frac{5S}{m}\right)^{1/2}\left[S^{-1}\sum_{l>0}l_1^4D^2g(r^{0l})\right.\right.\\&\qquad\left.\left.-\tfrac{1}{2}\{S^{-1}\sum_{l>0}l_1^2Dg(r^{0l})\}^2\right]\right\}(e_{11}^2+e_{22}^2+e_{33}^2)\\&+\frac{a^4}{8}\left\{S_{12}^{(0)}+\frac{3\hbar}{8}\left(\frac{5S}{m}\right)^{1/2}\left[S^{-1}\sum_{l>0}l_1^2l_2^2\,D^2g(r^{0l})\right.\right.\\&\qquad\left.\left.-\tfrac{1}{2}\{S^{-1}\sum_{l>0}l_1^2\,Dg(r^{0l})\}^2\right]\right\}(e_{11}e_{22}+e_{22}e_{33}+e_{33}e_{11})\\&+\frac{a^4}{4}\left\{S_{12}^{(0)}+\frac{3\hbar}{8}\left(\frac{5}{mS}\right)^{1/2}\sum_{l>0}l_1^2l_2^2D^2g(r^{0l})\right\}(e_{12}^2+e_{23}^2+e_{31}^2)+\ldots..\end{aligned}$$
$$. \quad . \quad . \quad (11)$$

Here $g(r^{0l})\equiv[3D\phi(r)+r^2D^2\phi(r)]_{r=r^{0l}}$, $S=\sum_{l>0}g(r^{0l})$, and the $S^{(0)}$ are the static lattice energy terms, as obtained by previous writers (e.g. Born 1940):

$$S_1^{(0)}=\sum_{l>0}l_1^2\,D\phi(r^{0l}),$$
$$S_{11}^{(0)}=\sum_{l>0}l_1^4\,D^2\phi(r^{0l}),$$
$$S_{12}^{(0)}=\sum_{l>0}l_1^2l_2^2D^2\phi(r^{0l}).$$

On the other hand, we have the relation from elasticity theory, valid for lattice structures of cubic symmetry:

$$\begin{aligned}F/Nv=f_0&-(p/2)(e_{11}+e_{22}+e_{33})+(c_{11}/8)(e_{11}{}^2+e_{22}{}^2+e_{33}{}^2)\\&+(c_{12}/4)(e_{11}e_{22}+e_{22}e_{33}+e_{33}e_{11})\\&+(c_{44}/2)(e_{12}{}^2+e_{23}{}^2+e_{31}{}^2)+\ldots\end{aligned}\qquad (12)$$

Here $v=\gamma a^3$ is the volume per molecule, and for the lattices here considered the γ values are: simple cubic, $\gamma=1$; face-centred cubic, $\gamma=2$; and body-centred cubic, $\gamma=4$. f_0 is the free energy density at absolute zero, p is the pressure; c_{11}, c_{12}, and c_{44} are the three elastic constants for a crystal of cubic symmetry. Dividing eqn. (11) by v and comparing coefficients with (12), explicit formulae for the quantities of interest at absolute zero are obtained. Adopting the Lennard-Jones potential function and writing it in the form

$$\phi(r)=4\epsilon[(\sigma/r)^{12}-(\sigma/r)^6], \qquad (13)$$

we have each of these quantities as functions of certain lattice sums involving inverse powers of $|\mathbf{r}^{0l}|$. Values of these lattice sums have been tabulated (Misra 1940, Barron 1953); we use them in the static lattice energy terms, but consider only nearest and next nearest neighbour terms in the Θ_∞ contributions. Numerically this restriction makes little difference (less than 4%), since terms arising from the dynamical matrix are insensitive to more than the first few rings of neighbours. We use reduced units (the reducing parameters ϵ and σ appearing in the force law (13)) to express the results, writing for the case of the face-centred cubic structure $V^*=(v/\sigma^3)$, $T^*=(kT/\epsilon)$, $p^*=(\sigma^3p/\epsilon)$, $\Lambda^*=h/\sigma\sqrt{(m\epsilon)}$, $c_{ij}{}^*=(\sigma^3c_{ij}/\epsilon)$, $a=\sigma(V^*/2)^{1/3}$, and obtain, finally, the following expressions:

$$U_0{}^*=\frac{f_0}{N\epsilon}=\frac{6\cdot066}{V^{*4}}-\frac{14\cdot454}{V^{*2}}+\frac{2\cdot291\Lambda^*}{V^{*7/3}}R^{1/2}; \qquad (14)$$

$$p^*=\frac{24\cdot26}{V^{*5}}-\frac{28\cdot91}{V^{*3}}+\frac{0\cdot3822\Lambda^*}{V^{*10/3}}\frac{(59\cdot96-16\cdot0V^{*2})}{R^{1/2}}; \qquad (15)$$

$$\begin{aligned}c_{11}{}^*=&\frac{171\cdot2}{V^{*5}}-\frac{123\cdot0}{V^{*3}}+\frac{1\cdot146\Lambda^*}{V^{*10/3}}R^{1/2}\\&\times\left[8\cdot275\left(\frac{19\cdot40-3\cdot320V^{*2}}{R}\right)-\frac{1}{18}\left(\frac{59\cdot96-16\cdot0V^{*2}}{R}\right)^2\right];\end{aligned} \qquad (16)$$

$$\begin{aligned}c_{12}{}^*=&\frac{84\cdot25}{V^{*5}}-\frac{54\cdot11}{V^{*3}}+\frac{1\cdot146\Lambda^*}{V^{*10/3}}R^{1/2}\\&\times\left[8\cdot275\left(\frac{9\cdot625-1\cdot563V^{*2}}{R}\right)-\frac{1}{18}\left(\frac{59\cdot96-16\cdot0V^{*2}}{R}\right)^2\right];\end{aligned} \qquad (17)$$

$$c_{44}{}^*=\frac{84\cdot25}{V^{*5}}-\frac{54\cdot11}{V^{*3}}+\frac{9\cdot483\Lambda^*}{V^{*10/3}}\frac{(9\cdot625-1\cdot563V^{*2})}{R^{1/2}}; \qquad (18)$$

$$c_{44}{}^*-c_{12}{}^*=\frac{0\cdot06367\Lambda^*}{V^{*10/3}}\frac{(59\cdot96-16\cdot0V^{*2})^2}{R^{3/2}}; \qquad (19)$$

where $$R\equiv4\cdot283-2\cdot00V^{*2}.$$

The use of the reduced units (de Boer 1940), where the parameters ϵ and σ are experimentally determinable from gaseous phase measurements, facilitate comparison with experimental results and in particular with the previous theoretical work of de Boer and Blaisse (1948).

Fig. 1

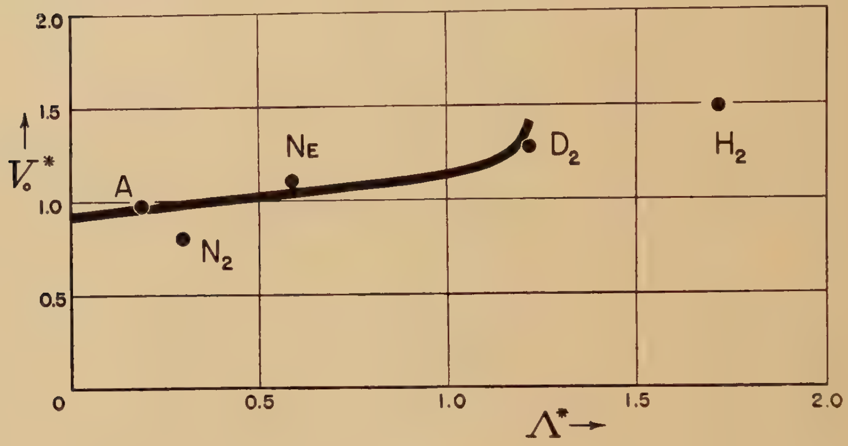
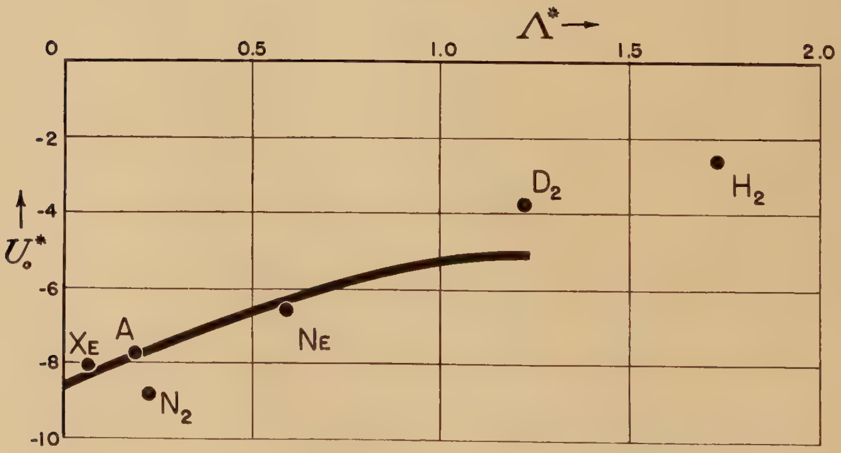


Fig. 2



§ 3

Figures 1 and 2 are after de Boer and Blaisse. Equation (15), with $p^*=0$, becomes a relation between V_0^*, the equilibrium volume at zero temperature, and the parameter Λ^*. Corresponding values are shown in fig. 1 as the smooth curve, while the points represent experimental values for the inert gas solids. Λ^* may be interpreted as the reduced de Broglie wavelength of relative motion for a substance fulfilling certain restrictions, and is a measure of the influence of specifically quantum effects on the behaviour of the substance. When corresponding values of V_0^* and Λ^* so obtained are substituted in eqn. (14), the curve for U_0^*, the latent heat of vaporization at absolute zero, as a function of Λ^* is the result as shown in fig. 2.

There is reasonable agreement to within the neighbourhood of the Λ^* value for deuterium, but the calculations break down for larger Λ^* values. The V_0^*–Λ^* curve passes through a maximum Λ^* value of 1·22 and then curves back rapidly toward the V_0^* axis, giving $\Lambda^*=0$ at $V_0^*=1{\cdot}46$. This behaviour is due to the variation of the factor $(4{\cdot}283-2{\cdot}0\ V^{*2})^{1/2}$ (essentially the volume dependence of Θ_∞^*); for reduced volumes greater than 1·46 Θ_∞^* is imaginary. This is the same sort of difficulty which occurred in the calculations of de Boer and Blaisse, where a Θ_0 computed from theoretical values of elastic constants was used for the zero point energy. One would expect, in the light of recent developments in the theory of the ideal crystal (Born 1951) that such difficulties are inherent in any approach based on the conventional ‘ semi-classical ’ lattice dynamics ; the zero point vibrations are so large for the solid elements with a very high Λ^* value (H_2, ^{4}He, ^{3}He) that the dynamical assumption of small vibrations may be no longer valid.

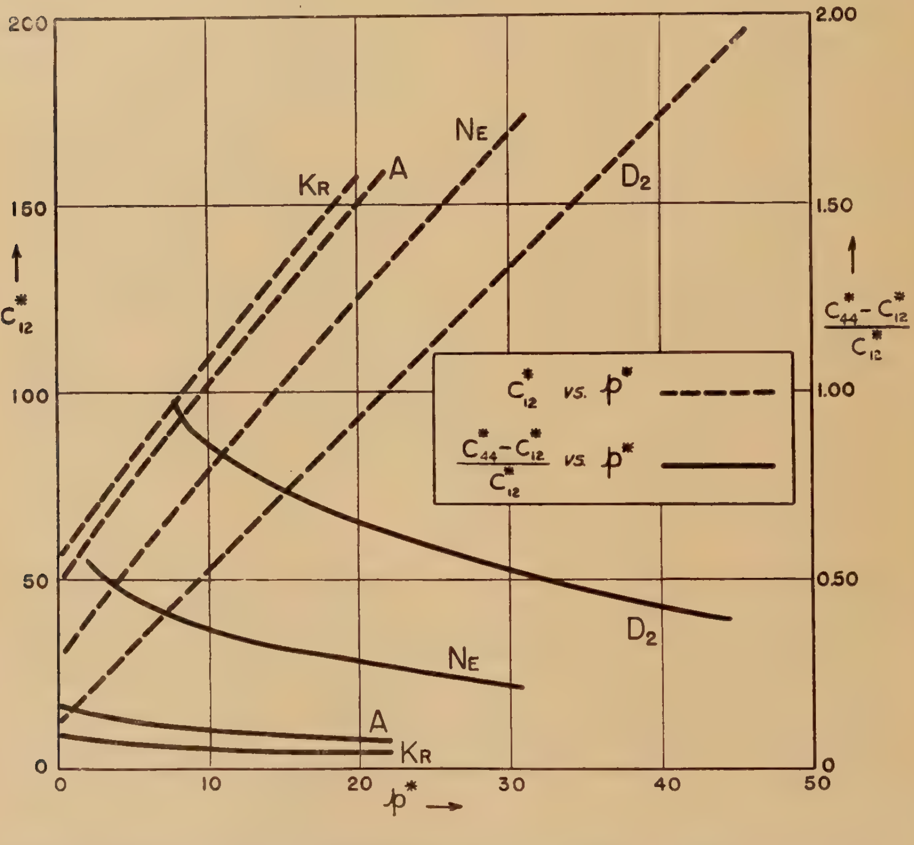
The comparison of theoretical with experimental values of V_0^* and U_0^* is not a very sensitive test for a zero point energy expression, partly due to the uncertainty in the experimental values themselves. The agreement obtained using Θ_∞ seems to be as good as that obtained by any previous calculations, and has the advantage of simplicity insofar as the actual numerical calculations are concerned.

§ 4

Of greater interest, perhaps, is the fact that the present theory yields an explicit expression for the elastic constants at absolute zero, consisting of the usual ‘ static ’ term, as obtained by Voigt and many others, plus an additional contribution due to the zero point energy term occurring in the free energy. This additional contribution is such, assuming that the zero point energy is correctly taken into account by the use of Θ_∞, as to destroy the validity of the Cauchy relation $c_{12}=c_{44}$ for the cubic lattices. Theoretical calculations of the equation of state and elastic constants at high temperatures have been made for simple monatomic substances crystallizing in the body-centred cubic (Born 1939) and face-centred cubic (Bradburn 1943) structures ; it was emphasized that for any finite temperature T the Cauchy relations no longer necessarily hold. It may not, however, be generally recognized that at absolute zero the presence of zero point energy may be sufficient to vitiate the relations.

In fig. 3 theoretical values of the reduced elastic constant c_{12} for the face-centred cubic lattice are plotted as functions of reduced pressure p^* for various values of Λ^*. The Λ^* values correspond to the inert gas solids A, Kr, Ne and D (dotted curves). The relative difference $(c_{44}^{*}-c_{12}^{*})/c_{12}^{*}=(c_{44}-c_{12})/c_{12}$ is plotted against pressure on the same figure for the same elements (solid curves). For example, the theory predicts that for the element Kr, at 0°K (physically, at temperatures low enough that the magnitude of contributions from the $(T/\Theta)^4$ and succeeding terms in the free energy expression are negligible), c_{12} has a value of $(3{\cdot}31)(10^{10})$ dyne/cm², while $c_{44}=(3{\cdot}54)(10^{10})$ dyne/cm², a relative deviation of approximately 7%. For Ne, at zero temperature

Fig. 3



and a pressure of 450 atmospheres, the relative deviation is about 54%, with $c_{12}=(0{\cdot}916)(10^{10})$ dyne/cm². Provided the elastic constant measurements are feasible, such differences should be detectable. The experimental difficulties are great, since the usual elasticity techniques must be combined with low temperature technique; there seem to be at present no measurements available for the substances to which the present theory is applicable (but see Barker *et al.* 1953). It is of interest to point out that in experiments on solid helium carried out at the Clarendon Laboratory Dugdale and Simon (1953) succeeded in determining the bulk modulus $K=(c_{11}+2c_{12})/3$ as a function of temperature and volume, and found a constant value at constant volume for temperatures such that $T/\Theta<0{\cdot}09$. In other words, for solid helium the range of temperatures such that only the static lattice energy and zero point energy terms of the free energy are appreciable is accessible experimentally. The solid elements Kr, A, Ne, etc. have Θ values greater than that of He ; it is thus conceivable that the predictions of this theory are capable of verification by measurements carried out at moderately low temperatures.

§ 5

Since the zero point energy makes a contribution to the magnitude of the elastic constants a deviation of the measured value from the usual theoretical value (calculated from the static lattice energy only) affords a quantitative test for any zero point energy expression (Simon 1934, Megaw and Simon 1936, Megaw 1939). Considering in particular the bulk modulus, for it is the most easily measured, we write

$$K=K_L+K_z, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (20)$$

K_L being the static lattice energy part and K_z the zero point energy part. We note that at absolute zero the isothermal and adiabatic moduli of elasticity coincide, so that there is no need to distinguish between them here. For the face-centred cubic lattice, in reduced units :

$$K_L{}^*=\frac{\sigma^3 K_L}{\epsilon}=\frac{113{\cdot}2}{V^{*5}}-\frac{77{\cdot}07}{V^{*3}} ; \quad . \quad . \quad . \quad . \quad . \quad (21)$$

$$K_z{}^*=\frac{1{\cdot}146\Lambda^*}{V^{*10/3}}R^{1/2}\left[\frac{106{\cdot}6-17{\cdot}83V^{*2}}{R}-\frac{1}{18}\left(\frac{59{\cdot}96-16{\cdot}0V^{*2}}{R}\right)^2\right],$$
$$R=4{\cdot}283-2{\cdot}0V^{*2}. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (22)$$

For argon at a pressure of 1800 atmospheres : $K_z/(K_L+K_z)=0{\cdot}076$; for neon at 5650 atmospheres : $K_z/(K_L+K_z)=0{\cdot}52$. No numerical values are given for K_L and K_z, since such values are sensitive to the form of the theoretical potential law adopted. For example, the potential $\phi(r)=ae^{-r/\alpha}-br^{-6}$ is preferable on theoretical grounds to the Lennard-Jones force law and may be expected to lead to somewhat different magnitudes for the c_{ij} and K, though reproducing quantitatively the proportional influence of static lattice and zero point energy terms. Pending a further investigation based on the use of different potential laws, we have here limited the quantitative considerations to the relative effect of the two energy contributions.

Provided, then, that experiments on the bulk modulus can be carried out in the ‘ plateau ’ region, where K is constant before the temperature dependent terms become effective and the value begins to drop with rising temperature, measurements on the lighter inert gases at medium pressures should give important information on the question of zero point energy and its influences on the elastic properties of the solid.

§ 6

The question of the validity of the Cauchy relations is important in the following connection: one of the usual criteria for the existence of central forces is the fulfilment of the Cauchy relations. The experimental fact that in metals the relations are generally not fulfilled is taken as confirmation of the supposition that metallic binding forces are chiefly non-central, a supposition which is of course amply substantiated by theoretical considerations. Any deviation due to the presence of zero point energy one would expect to be negligible in the case of metals, for experiment indicates that with the exception of the lightest solids the influence of zero point energy on the properties is negligible. It is, however, precisely in the case of these exceptional, inert gas type solids that the assumption of central forces has the strongest theoretical basis. Should experiment show that for these 'ideal' monatomic solids the Cauchy relations are not valid, this would not necessarily invalidate the central force hypothesis; the present calculations are based on such an assumption and yet indicate that in general such a deviation could be expected due to the influence of zero point energy.

ACKNOWLEDGMENTS

The author is indebted to Messrs. C. Domb, R. W. Hill, and T. H. K. Barron for helpful discussion; to Dr. F. K. Elder Jr. for preparation of the diagrams; and to the Rhodes Trust for financial assistance.

REFERENCES

BARKER, DOBBS, and JONES, 1953, *Phil. Mag.*, **44**, 1182.

BARRON, 1953, unpublished results; work carried out at the Clarendon Laboratory, Oxford.

BORN, 1939, *J. Chem. Phys.*, **7**, 591; 1940, *Proc. Camb. Phil. Soc.*, **36**, 160; 1951, *Festschrift*, Akademie der Wissenschaften in Göttingen (Mathematisch-Physikalische Klasse).

BRADBURN, 1943, *Proc. Camb. Phil. Soc.*, **39**, 113.

DE BOER, 1940, *Contribution to the Theory of Compressed Gases* (Amsterdam).

DE BOER and BLAISSE, 1948, *Physica*, **14**, 139.

DOMB and SALTER, 1952, *Phil. Mag.*, **43**, 1083.

DUGDALE and SIMON, 1953, *Proc. Roy. Soc.* A, **218**, 291.

MEGAW, 1939, *Phil. Mag.*, **28**, 142.

MEGAW and SIMON, 1936, *Nature, Lond.*, **138**, 244.

MISRA, 1940, *Proc. Camb. Phil. Soc.*, **36**, 173.

SIMON, 1934, *Nature, Lond.*, **133**, 529.

STAKGOLD, 1950, *Q. Appl. Math.*, **8**, 169.

VOIGT, 1928, *Lehrbuch der Kristallphysik* (Leipzig).

XLIV. *The Simon Melting Equation*

By L. SALTER*
The Clarendon Laboratory, Oxford†

[Received January 18, 1954]

ABSTRACT

The Simon melting equation $p_m/a=(T_m/T_0)^c-1$ is derived assuming the validity of the Grüneisen equation of state and the Lindemann melting formula along the melting line. A relation between c and Grüneisen's γ is derived and discussed in view of available experimental data. Poor agreement is found in the case of the alkali metals, the agreement for argon being satisfactory. A consequence of possible geophysical interest arising from the theory is discussed. The detailed data available for solid helium are used to study the assumptions underlying the theory.

§ 1

THE purpose of this paper is to point out a relation between three well known equations connected with the solid state :

The Grüneisen equation of state

$$p=-\frac{dU_0}{dV}+\frac{\gamma}{V}(U-U_0), \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (1)$$

the Lindemann melting formula

$$\frac{mV_m{}^{2/3}\theta^2}{T_m}=\text{const.}, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (2)$$

and the Simon melting equation

$$\frac{p_m}{a}=\left(\frac{T_m}{T_0}\right)^c-1. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (3)$$

In (1) p is the pressure, U_0 is the internal energy at absolute zero, $U-U_0=\int_0^T C_v\,dT$ is the thermal energy, θ (not necessarily the Debye or Einstein θ) is a function $\theta(V)$ of volume only such that $C_v=f(T/\theta)$, and $\gamma=-d\ln\theta/d\ln V$ is Grüneisen's 'constant'. In (2), θ is again a characteristic temperature of some kind, V_m and T_m are simultaneous volume and temperature values along the melting line, and m is the atomic mass. In the Simon equation (3) p_m and T_m are the melting curve values of pressure and temperature, T_0 is the triple point temperature, and a (essentially the internal pressure) and c are constants.

---

* Communicated by Professor F. Simon, F.R.S.

† Now at Wabash College, Crawfordsville, Indiana, U.S.A.

Now eqn. (1), though originally derived by Grüneisen in an extension of the Mie theory of solids and based on a particular model, is in a sense a thermodynamic relation. That is, given the relation $C_v = f(T/\theta)$, with θ a function of volume only, eqn. (1) and the equally important relation

$$\frac{\alpha}{\kappa} = \frac{\gamma C_v}{V} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (4)$$

(κ the compressibility at constant temperature, α the volume expansion coefficient) follow at once by simple thermodynamic reasoning (see, for example, Davies 1952). Theoretically, the restrictive condition on C_v for a Grüneisen solid implies that the 3N eigenfrequencies (for a monatomic solid) of the dynamical matrix are independent of temperature and vary identically with volume. Grüneisen (1926) has shown that experiments on a number of solids are adequately described by the theory. (It has recently been shown by Bijl and Pullan (in print *Phil. Mag.*) that Grüneisen's relation breaks down at temperatures below θ ; we are here, however, concerned mainly with much higher temperatures only.)

The Lindemann equation (2), while originally derived (Lindemann 1910) on the basis of a simple model of the solid and a particular picture of the melting process, is undoubtedly of wider validity ; it finds justification in that a number of substances are found to satisfy the formula at the normal melting point, while recently Dugdale and Simon (1953) have found that solid helium satisfied eqn. (2) over a wide portion of the melting curve. Values of the constant vary somewhat from one type of solid to another, but for the majority of solids the constants lie within a small range of variation.

The melting curve eqn. (3) was proposed by Simon (1929) after a careful study of experimental data ; it is found that for all substances heretofore studied an a and a c can be determined which will make $\ln (p_m + a)$ and $\ln T_m$ vary linearly. The equation has been very useful as a means of representing experimental results for a wide variety of substances over varying melting temperature and pressure ranges, and in the planning of further experiments (e.g., the use of solid helium as a ‘ model ’ substance (Simon 1937, 1952 a)). It is somewhat surprising, therefore, that a relation of such wide experimental validity should have, apparently, little theoretical foundation. Of the few theoretical derivations which have been published, that of Domb (1951) as improved by de Boer (1952) is the most successful ; it is based on an adaptation of the Lennard-Jones and Devonshire (1939) model of the melting process. The results are restricted in validity by the assumption of high pressures and ‘ classical ’ substances (negligible zero point energy, $T_m/\theta \gg 1$).

We shall show that eqn. (3) follows in an elementary fashion from (1) and (2) on the basis of certain reasonable assumptions.

§ 2

First, we assume that the Grüneisen equation of state holds along the melting curve. We write, then, for (1):

$$p_m = -U_0'(V_m) + \frac{\gamma}{V_m}(U-U_0)_{T_m, V_m}. \quad . \quad . \quad . \quad . \quad (5)$$

Obviously, by eliminating V_m between (5) and the Lindemann equation (2) we will be led to a melting curve equation of some form. It is first necessary to consider the physical significance of the parameter a occurring in the Simon relation. Simon (1952 b) has put forth strong evidence that a is related to the internal pressure $(\partial U/\partial V)_T$ of the solid; in particular, $a=(\partial U/\partial V)_{T=0}$. For, writing eqn. (3) at $T_m=0$ (assuming that this extrapolation is valid)

$$p_m = -a. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (6)$$

On the other hand, we have the thermodynamic relation

$$\left(\frac{\partial U}{\partial V}\right)_T = T\left(\frac{\partial p}{\partial T}\right)_V - p \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (7)$$

and in particular, on the melting curve intercept at $T_m=0$

$$\left(\frac{\partial U}{\partial V}\right)_{T_m=0} = -p_m. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (8)$$

Comparing (6) and (8) we are led to the relation

$$a = \left(\frac{\partial U}{\partial V}\right)_{T_m=0}. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (9)$$

We apply these considerations to the Grüneisen equation (5); by definition $(\partial U/\partial V)_{T=0} = U_0'(V)$, thus

$$a = U_0'(V_m). \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (10)$$

We assume in all that follows that $U_0'(V_m)$ is constant along that portion of the melting line corresponding to low melting pressures. If melting in the vicinity of the triple point (essentially atmospheric pressure) takes place at volumes near the inflection point on the $U_0(V)$ curve, this should be a reasonable assumption. Naturally, at high melting pressures the volume will be decreased and $U_0'(V_m)$ will vary, but this variation in the theoretical a would be completely masked by the large value of p_m. We note that the identification (11) was the essential improvement made by de Boer (1952) on Domb's derivation of (3). Now eqn. (5) becomes

$$p_m + a = \frac{\gamma}{V_m}(U-U_0). \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (11)$$

To proceed further we must assume some definite functional dependence of $U-U_0$ on V_m and T_m; the limiting high temperature expression $3NkT_m$ should suffice for most solids (we have in mind throughout monatomic solids, for the extension is obvious), the exceptional case of solid helium being reserved for later discussion. If we now assume that γ is constant, so that we may write

$$\theta = b(h/km^{1/2})V^{-\gamma} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (12)$$

b a constant determined by the lattice structure and the interatomic potential, then, substituting in (11) and eliminating V_m between

$$p_m+a=3NkT\gamma/V_m, \quad \theta=b(h/k_m{}^{1/2})V^{-\gamma},$$

and $m\,V_m{}^{2/3}\,\theta^2/T_m=h^2B/k$ (B dimensionless), we find

$$p_m+a=3Nk\gamma\left(\frac{Bk}{b^2}\right)^{c-1}T_m{}^c \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (13)$$

with

$$c=\frac{6\gamma+1}{6\gamma-2}. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (14)$$

Applying this at the triple point T_0, where p_m may be neglected in comparison with a,

$$a=3Nk\gamma\left(\frac{Bk}{b^2}\right)^{c-1}T_0{}^c \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (15)$$

and dividing (13) by (15) we establish $p_m/a=(T_m/T_0)^c-1$, the Simon formula.

This derivation, then, connects the melting curve parameter c with Grüneisen's γ, the latter being determined by the form of the interatomic potential energy function and the dynamical behaviour of the lattice. We note that it has been assumed implicitly in the foregoing, in addition to the constancy of γ and $U_0'(V_m)$ along the melting curve, that the characteristic temperature θ occurring in (1) and (2) is the same function, at least in so far as volume dependence is concerned.

By means of the Clausius–Clapeyron equation we obtain, differentiating (3) and using (15)

$$\frac{dp_m}{dT_m}=\left(\frac{\Delta S_m}{\Delta V_m}\right)=\frac{ac}{T_0}\left(\frac{T_m}{T_0}\right)^{c-1}=3Nk\gamma\left(\frac{6\gamma+1}{6\gamma-2}\right)\left(\frac{BkT_m}{b^2}\right)^{c-1} \quad . \quad (16)$$

and at the triple point T_0

$$\left(\frac{dp_m}{dT_m}\right)_{T_0}=\left(\frac{\Delta S_m}{\Delta V_m}\right)_{T_0}=3Nk\gamma\left(\frac{6\gamma+1}{6\gamma-2}\right)\left(\frac{BkT_0}{b^2}\right)^{c-1}. \quad . \quad . \quad (17)$$

Equations (16) and (17) give further relations involving the γ of the solid phase and experimentally determinate quantities associated with melting.

§ 3

Professor Simon has pointed out to the writer that the relation (14) connecting c and γ is of interest in the following regard. The condition that the entropy should increase along the melting curve with rising pressure, or alternately, that melting of a substance by the application of adiabatic compression alone be impossible, may be reduced to the demand that T_m/θ should increase with p_m (Dugdale and Simon 1953). The present theory enables us to express this condition in terms of γ for the substance. For

$$\frac{d}{dp_m}\left(\frac{T_m}{\theta}\right)=\frac{T_m}{ac}\left(\frac{T_0}{T_m}\right)^c\frac{d}{dT_m}\left(\frac{T_m}{\theta}\right)$$

$$=\left(\frac{3\gamma-2}{6\gamma-2}\right)\frac{1}{ac}\left(\frac{T_0}{T_m}\right)^c\left(\frac{T_m}{\theta}\right)\geqslant 0 \quad \text{for} \quad \gamma\geqslant 2/3. \quad . \quad . \quad (18)$$

The limiting value $\gamma=2/3$ is, somewhat artificially, the limiting theoretical value for $V\rightarrow\infty$ (i.e., the perfect gas), as has been pointed out to the writer by Dr. C. Domb. Here γ for a perfect gas is defined by the relation (4), or equivalently, by $\gamma=-(\partial \ln T/\partial \ln V)_S$. Using (14), the restriction $\gamma>2/3$ may be expressed as $c<5/2$. The only metals for which experimental c and γ values are available are the alkali metals ; without exception the experimental values available satisfy $\gamma>2/3$ but $c>5/2$. As stated below, there is some uncertainty in the γ values, so that the implied inconsistency in eqns. (14) and (18) may be only apparent. Pending further investigation of the question, it may be pointed out that the entropy of solid sodium has been shown from experimental data to increase along the melting curve (Simon 1937). The case of greatest interest geophysically is iron ; Grüneisen (1926) found by calculation, using eqn. (4), $\gamma=1{\cdot}60$, so that the condition for entropy increase along the melting line is satisfied.

§ 4

Broadly speaking, a relation between γ and c of the form (14) does seem plausible in view of available experimental data. Metals, which have low γ values, have values of c ranging from 2 to 4 (but again, we have melting curves for only the most compressible metals). The inert gas solids, on the other hand, have relatively high γ's (if we take the lone value for argon as representative) and $c\simeq 1{\cdot}5$. Quantitative checking of the theory is made difficult by the fact that for most substances γ must be computed indirectly from the experimental data ; this may be done in several different ways and the values so obtained are not always in good agreement. At the same time, experimental difficulties in some cases preclude melting curve measurements of sufficient accuracy to determine unique values for c and a.

In table 1 are given experimental values of γ, the theoretical values of c computed using eqn. (14), and the experimental values of c.

The agreement in the case of the alkali metals is distinctly poor, and we conclude that for these highly compressible metals the assumptions underlying the present theory are not fulfilled. Nevertheless there seems to be a correlation in relative magnitude between experimental and theoretical c's (only Na is out of line). We have used Grüneisen's calculated values of γ, computed through eqn. (4), as being the most reliable in the absence of direct determinations. These depend critically on compressibility values ; in fact, it may well be that for these metals the theoretical calculations of γ must be altered in accord with finite deformation theories (Truesdell 1952).

It might be expected that the substances most likely to fulfill the assumptions would be the heavier inert gas solids : A, Kr, and Xe. Unfortunately, the only γ value available (with the exception of the direct measurements for solid He as discussed below) is for argon, as listed in table 1. The good agreement between computed and theoretical values of c in this case is blighted somewhat in that the value $\gamma=4{\cdot}5$ is suspect as being too high (private communication from Professor Simon). However, correction to the more probable value 3·5 only betters the agreement, giving $c=1{\cdot}16$ in agreement with Robinson's measured value. Experimental γ measurements for these rare gas solids would be of great interest, not only in checking the present theory, but because γ as a function of V is very sensitive to the form of the interatomic potential energy curve.

Table 1

Substance	γ	c_{theor}	c_{exptl}
Na	1·25 (1)	1·55	3·56 (7)
K	1·34 (1)	1·50	4·53 (7)
Rb	1·48 (1)	1·44	4·2 (7)
Cs	1·29 (1)	1·52	4·75 (2)
A	4·5 (3)	1·12	1·16 (4) 1·228 (5) 1·478 (6)

References : (1) Grüneisen, 1926, *Handb. der Physik*, **10.**
(2) Simon and Glatzel, 1929, *Zeits. anorg. allg. Chem., Bd.* 178, 309.
(3) Simon and Kippert, 1928, *Z. f. phys. Chemie*, **135,** 113.
(4) Robinson, 1952, *Thesis*, Oxford.
(5) Simon, Ruhemann and Edwards, 1930, *Z. f. phys. Chemie*, B, **6,** 331.
(6) Bridgman, 1935, *Proc. Am. Acad. Arts and Sciences*, **70,** 1.
Note : Of the three c values listed for argon, that of Robinson is probably the most accurate, as his experiments covered a wider melting pressure range.
(7) Data by Bridgman, 1949, *The Physics of High Pressures*, London. c values computed by Dr. J. A. W. Huggill. The values for lithium have been omitted as only a rather impure sample was available.

§5

Equation (14) gives a relation between a and T_0 for a particular solid, involving the constants c, b and B. On theoretical grounds (de Boer 1948) we can expect that b and B depend only on the lattice structure and on the shape of the potential energy curve ; by ‘ shape ’ is meant that the potential function can be written in the form $\phi(r)=\epsilon g(r/\sigma)$, where $g(x)$ is a universal function specifying the form of the potential, ϵ and σ being reducing parameters characteristic of the particular solid which specify the scale. The properties of the heavier inert gas solids seem to be well described theoretically through the use of such a ‘ reduced ’ potential function ; it is known that they crystallize in the same structure, the face centered cubic. Re-examining our equations in the light of these dimensional considerations we write for the constant b occurring in eqn. (12)

$$b=\frac{\beta\epsilon^{1/2}}{\sigma}(N\sigma^3)^{\gamma},$$

where now β is dimensionless and should, for corresponding solids, depend only upon the lattice structure. We can express the equations in reduced form, using the variables $V^*=V/N\sigma^3$, $p^*=p\sigma^3/\epsilon$, $T^*=kT/\epsilon$, and $\Lambda^*=h/\sigma\surd(m\epsilon)$ (de Boer 1948) and derive as before

$$p_m^*+a^*=3\gamma\left(\frac{B}{\beta^2N^{2/3}}\right)^{c-1}(T_m^*)^c, \qquad \ldots\ldots \qquad (13')$$

with $c=(6\gamma+1)/(6\gamma-2)$ as before. Equation (15) becomes

$$a^*=3\gamma\left(\frac{B}{\beta^2N^{2/3}}\right)^{c-1}(T_0^*)^c. \qquad \ldots\ldots \qquad (15')$$

Now Clusius (1936) has determined the constant B for A, Kr, and Xe, finding the almost identical values $(1{\cdot}39)(10^{17})$, $(1{\cdot}40)(10^{17})$, and $(1{\cdot}41)(10^{17})$ respectively. Since these solids have the same lattice structure, we expect the β's and γ's to be identical. Hence we can write (15′) in the form

$$a^*=K(T_0^*)^c, \qquad \ldots\ldots \qquad (19)$$

K and c the same constants for each of the corresponding solids. These theoretical considerations lead us to expect that experimental values of c will be very nearly the same for these three rare gas solids, and (using Robinson's a and c values for argon, known triple point temperatures, and values of ϵ and σ determined from gaseous phase experiments) that a for Kr and Xe will be approximately 3900 and 4400 atmospheres respectively. Clusius (1940) has determined the melting curves for Kr and Xe, but over too small a pressure range to allow a unique determination of a.

Exactly the same considerations apply to eqn. (17); in reduced form

$$\left(\frac{\Delta S^*}{\Delta V^*}\right)_{T_0^*}=\frac{1}{T_0^*}\left(\frac{\Delta H^*}{\Delta V^*}\right)=3\gamma c\left(\frac{B}{\beta^2N^{2/3}}\right)^{c-1}(T_0^*)^{c-1}, \qquad . \qquad (17')$$

or

$$\frac{1}{(T_0^*)^c}\left(\frac{\Delta H^*}{\Delta V^*}\right)_{T_0^*}=3\gamma c\left(\frac{B}{\beta^2N^{2/3}}\right)^{c-1}=\text{const.} \qquad \ldots\ldots \qquad (20)$$

Experimental values of ΔH and ΔV are available (Guggenheim 1950) and, using Robinson's value of c for argon, we get the following computed values of $(T_0^*)^{-c}(\Delta H^*/\Delta V^*)_{T_0^*}$ for A, Kr, and Xe: 12·0, 11·7, and 11·7 respectively. The fact that these values are very nearly the same confirms the idea that the heavier inert gases may be adequately described by a 'classical' reduced equation of state (de Boer and Blaisse 1948). The present theory, however, gives an expression for the magnitude of the constant in terms of other experimentally determinate quantities. Since no reliable experimental value of γ for any of these solids is available, we can only check for consistency. Taking Robinson's value for A, $c=1{\cdot}160$, we use eqn. (14) to get $\gamma=3{\cdot}56$. Using simultaneous experimental values for B, θ_D, and V_m (Clusius 1936) for A at the triple point, the number $3\gamma c(B/\beta^2N^{2/3})^{c-1}$ can be evaluated; the result is 12·7, as compared with the previously given 12·0 (determined from $\Delta H_m/\Delta V_m$). The same calculation, using instead the unreliable experimental value $\gamma=4{\cdot}5$, gives the result 16·2. Such good agreement of the two sides of eqn. (20) may be in part confirmation of the theory or else fortuitous ; a satisfactory test of the theory in this direction awaits accurate γ determinations.

§ 6

In the case of solid helium we are in possession of a fairly complete set of experimental data with which to check our assumptions, thanks to the work of Dugdale and Simon (1953). The specific heat at constant volume C_v was measured and the measurements can be represented by a universal function $f(T/\phi)$, with ϕ a function of volume only. $f(x)$ is not exactly a Debye function, however ; the equivalent Debye θ's were computed as a function of temperature for various densities. Further, the measurements permitted a direct calculation of γ by means of the fundamental definition $\gamma=-d\ln\phi/d\ln V$; γ is constant at approximately 2·4 over the greater portion of the volume range covered by the experiments, falling off gradually at lower volumes. The Lindemann relation was found to hold within the experimental error over the greater portion of the melting curve, not for ϕ, but for the equivalent Debye $\theta(V_m, T_m)$. If the function $\phi(V_m)$ is substituted in eqn. (2), the constant seems to vary steadily along the melting line. Writing (2) in the form

$$\frac{m\phi^2 V_m{}^{2/3}}{T_m}=\frac{h^2}{k}DT_m{}^n, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \qquad (21)$$

where now D is constant, the value $n=0{\cdot}26$ is found to represent the results of Dugdale and Simon. Thus our implicit assumption as to the identity of the two characteristic temperatures occurring in (1) and (2) is not fulfilled in the case of helium ; the modified Lindemann relation (21) must be used for calculations. Further, the following two modifications must be made : (*a*) the zero point energy, which up to now has been neglected, must be explicitly taken into account, as it plays a dominant part in the behaviour of helium in the condensed phase ; and (*b*) the energy expression $U-U_0\simeq 3NkT_m$ must be altered, since melting takes place at relatively low temperatures for helium.

The zero point energy is included in $U_0(V)$, and thus enters the Grüneisen equation through $U_0'(V)$. We can (in theory, at least) separate U_0 in the usual manner, writing $U_0=\Phi+U_z$. Here Φ is the static lattice energy ; Dugdale and Simon found that good agreement with London's theoretical calculation (London 1936) of Φ could be obtained if U_z was represented by the Debye expression $(9/8)Nk\phi$. The magnitude of ϕ was chosen so as to coincide insofar as possible with the equivalent Debye θ at $T=0$. Hence eqn. (5) becomes

$$p_m=-\Phi'(V_m)+\frac{9}{8}\frac{Nk\gamma\phi}{V_m}+\frac{\gamma}{V_m}(U-U_0)_{T_m, V_m}. \quad . \quad . \qquad (22)$$

Now $\phi(V_m)$ is connected with T_m through the modified Lindemann eqn. (21) and thus should vary strongly with temperature on the melting line. We tentatively assign the role of the parameter a to the quantity $\Phi'(V_m)$, assuming that the melting volumes at low pressure are in the vicinity of the inflection point on the $\Phi(V)$ curve. Writing

$$a=\Phi'(V_m), \qquad (23)$$

eqn. (22) becomes

$$p_m+a=\frac{9}{8}\frac{Nk\gamma\phi}{V_m}+\frac{\gamma}{V_m}(U-U_0). \qquad (24)$$

The question of the proper theoretical form of $U-U_0$ in terms of T_m and V_m is a difficult one, for the lattice dynamical theory of solid helium is still in an unsettled state. We can expect that $U-U_0$ for helium in the temperature range covered by these experiments may be adequately represented by an expression of the form

$$U-U_0\simeq 3NkT \sum_{l=1} M_{2l+1}(T/\phi)^{2l+1}, \qquad (25)$$

and as a crude approximation, valid only at the lowest temperatures, we neglect all but the first term ; we take $M_3=\pi^4/15$ (the Debye expression) as giving sufficient account of the correct coefficient.

We have then, approximately,

$$p_m+a=\frac{9Nk\gamma}{8}\frac{hb}{km^{1/2}}\left(\frac{Dk}{b^2}\right)^{q_1}T_m{}^{c_1}+3Nk\gamma\frac{\pi^4}{15}\left(\frac{Dk}{b^2}\right)^{q_2}\left(\frac{m^{1/2}k}{hb}\right)^3 T_m{}^{c_2} \qquad (26)$$

where

$$q_1=(1+\gamma)(c-1), \qquad c_1=(1+\gamma)(1+n)(c-1),$$

$$q_2=(3\gamma-1)(c-1), \quad \text{and} \quad c_2=3[1-\gamma(1+n)(c-1)]+1, \qquad (27)$$

after elimination of V_m. Substituting experimental values for the various parameters we find a melting curve equation

$$p_m+a\simeq 57{\cdot}7T_m{}^{1{\cdot}04}+(9{\cdot}33)(10^{-3})T_m{}^{2{\cdot}12}+\ldots \qquad (28)$$

We see that for all but the highest temperatures (when succeeding terms give additional contributions) the first term dominates and we get an equation of the form of (3), but with $c=1{\cdot}04$ as compared with the experimental value (Robinson 1952) 1·562. Naturally we should expect to be able to fit the melting curve using an equation of the form

$$p_m+a=K_1T_m{}^{c_1}+K_2T_m{}^{c_2}+\ldots,$$

for there are more parameters. The fact remains that the melting curve of helium is well described (with the possible exception of the extreme high pressure portion) by an equation of the form (3). We must conclude that the present considerations fail to account for this satisfactorily. It may well be that the general procedure set forth here is correct in essentials, but that the highly arbitrary step of writing $U_0=\Phi+U_z$, $U_z=(9/8)Nk\phi$ is incorrect. A correct microscopic theory of solid helium (based perhaps on Born's recent extension of lattice theory (Born 1951)) is at any rate necessary for further understanding of experimental results.

§ 7

In conclusion, it is emphasized that no mechanism or description of the melting process has been postulated ; indeed, this whole question has been by-passed through the assumption of the Lindemann relation from the beginning. Rather, these considerations suggest that eqn. (3) might be, theoretically, of the same degree of validity as the well known eqns. (1) and (2), following as it does quite directly from them under certain assumptions. These assumptions seem reasonable in theory ; the experimental evidence available for the alkali metals and for helium, however, does not wholly confirm them. The evidence is not complete for the heavier inert gas solids and predictions have been made from this heory which may serve, when further experiments become possible, to reject or further verify this explanation of the validity of the Simon melting equation.

ACKNOWLEDGMENTS

The author is indebted to Prof. F. E. Simon for his interest and numerous helpful discussions, to Dr. C. Domb and Mr. T. H. K. Barron for advice and assistance in preparing the manuscript, to Dr. J. A. W. Huggill for performing the calculations (previously unpublished) of c from Bridgman's data included in table 1, and to the Rhodes Trust for financial assistance.

REFERENCES

BORN, 1951, *Festschrift zur Feier des 200 jährigen Bestehens der Akademie der Wissenschaften in Göttingen.*

CLUSIUS, 1936, *Z. Phys. Chem.* B, **31,** 459.

CLUSIUS and WEIGAND, 1940, *Z. Phys. Chem.* B, **46,** 1.

DAVIES, 1952, *Phil. Mag.*, **43,** 472.

DE BOER, 1952, *Proc. Roy. Soc.* A, **215,** 4.

DE BOER and BLAISSE, 1948, *Physica*, **14,** 139, 149, 520.

DOMB, 1951, *Phil. Mag.*, **42,** 1316.

DUGDALE and SIMON, 1953, *Proc. Roy. Soc.* A, **218,** 291.

GRÜNEISEN, 1926, *Handbuch der Physik*, **10,** 1.

GUGGENHEIM, 1950, *Thermodynamics* (Second Ed.), p. 144 (Amsterdam : North Holland Publ. Co.).

LENNARD-JONES and DEVONSHIRE, 1939, *Proc. Roy. Soc.* A, **170,** 464.

LINDEMANN, 1910, *Phys. Z.*, **11,** 609.

LONDON, 1936, *Proc. Roy. Soc.* A, **153,** 576.

ROBINSON, 1952, *Thesis*, Oxford.

SIMON, 1937, *Trans. Faraday Soc.*, **33,** 65 ; 1952 a, *Low Temperature Physics* (London : Pergamon Press), p. 24 ; 1952 b, *L. Farkas Memorial Volume* (Jerusalem : Research Council of Israel), Special Publication No. 1, p. 37.

SIMON, RUHEMANN and EDWARDS, 1929, *Z. Phys. Chem.* B, **6,** 62 ; 1930, *Z. Phys. Chem.* B, **6,** 331.

TRUESDELL, 1952, *J. Rational Mech. Anal.*, **1,** 125.

XLV. *Spins of Odd–Odd Nuclei in the j–j Coupling Model: I*

By A. Hitchcock*
Cavendish Laboratory, Cambridge†

[Revised MS. received December 14, 1953]

Abstract

An attempt is made to justify the empirical rules for the spins of odd–odd nuclei recently proposed. It is found possible to do so for certain classes of nuclei, which happen to include nearly all those for which the experimental evidence is reliable and the rules obeyed. For one of the classes, however, it is necessary to make the approximation of assuming nuclear forces of infinite range.

§ 1. Introduction

The direct experimental measurements of spins of odd-odd nuclei are scanty, since they are necessarily limited to stable or long-lived nuclei. It is possible, however, to make more or less definite assignments in many cases as a result of studies of decay schemes and *ft* values. It is naturally tempting, in view of the succcesses of the spin–orbit coupling model, to see if the values so obtained have any simple interpretation on this basis. Empirical rules have been formulated by Nordheim (1951), who summarizes his result in these terms :

" (i) The individual configuration of neutrons and protons in odd–odd nuclei are the same as in odd–A nuclei with the same number of nucleons in the odd particle group.

(ii) If the odd neutron and proton belong to different Schmidt groups (i.e. $j_p=1\pm\frac{1}{2}$, $j_n=1\mp\frac{1}{2}$) their resultant spins will subtract.

(iii) If the odd neutron and proton belong to the same Schmidt group (i.e. $j_p=1\pm\frac{1}{2}$, $j_n=1\pm\frac{1}{2}$) their resultant spins will couple to a larger than minimum resultant . . . in many cases near the possible maximum (j_p+j_n)."

The strict form of rule (iii) $(I=j_p+j_n)$ will be referred to as (iii′). Detailed calculations to be reported later support rule (i) to a limited extent. The formulation above is chosen in order to account for the spins 3 and 4 of the sodium isotopes ^{22}Na and ^{24}Na, and these as it happens are two of the cases in which agreement with rule (i) is least good. However, the rule does seem to be generally in approximate agreement with the results of detailed calculations.

---

[*] Communicated by the Author.

[†] Now at Department of Atomic Energy, Industrial Group Headquarters, Risley, Warrington, Lancs.

Most of the possible states of an odd–odd nucleus have spins of the order (j_p+j_n), and in the absence of any special symmetry requirements such as those applying in even–even nuclei, we would expect spins of this order. Rule (ii) and rule (iii′), therefore, are more significant than rule (iii). Other things being equal, and accepting rule (i), it is clear that spin-dependent forces will favour parallel spins of proton and neutron, as in the deuteron, and so favour both (ii) and (iii′). But nuclear forces are predominantly non-spin-dependent, and it seems worthwhile to seek theoretical justification of the rules more searching than the remark just made. This is attempted in the following ; but we must first point out that the experimental evidence for the rules is in fact more limited than is claimed by the proposer.

Nordheim (1951), indeed, quotes a long list of spins, with shell assignments for the odd particles. Many of the latter seem a little arbitrary, and indeed, for $A>60$, there is, in general, sufficient competition between shells for almost any datum of the type available to be fitted into the scheme of the rules. We thus need only consider nuclei with $A<60$.

Now nearly all nuclei with $A<60$ fall into one of the classes detailed below, for which we shall justify the rule theoretically, and table 1 shows that those which do not, frequently do not satisfy the rule also ; especially when it is remembered that rule (i) has been tailored to fit the Na isotopes.

Table 1. Odd–Odd Nuclei, $A<60$, Showing Agreement with Rules (ii) and (iii′)

	Class A	Class B	Others
Rule obeyed	^{2}H, ^{10}B, ^{14}N	—	^{16}N, ^{22}Na, ^{24}Na ^{42}K, ^{50}V
Rule possibly obeyed	^{34}Cl, ^{38}K, ^{48}Sc	^{30}P, ^{32}P, ^{34}P, ^{18}F	^{12}B, ^{12}N, ^{38}Cl
Rule not obeyed	^{6}Li,* ^{36}Cl	—	^{26}Al, ^{40}K, ^{44}Sc, ^{46}Sc, ^{52}Mn, ^{48}V§
Insufficient evidence	—	—	^{20}F, ^{28}Al

* ^{6}Li is not a true exception. In this light nucleus the tensor forces must be considered. See Hitchcock (1952).

§ See Nussbaum, van Lieshout and Wapstra (1953).

§ 2. DETAILED CALCULATIONS

The two classes of nuclei mentioned above are :

A. 1 nucleon or 1 hole in both proton shell and neutron shell, protons and neutrons in the same shell.

B. 1 of the odd nucleons is in an $s_{1/2}$ shell, and there is 1 nucleon or 1 hole in the shell of nucleons of the other kind.

In Class A we are only able to justify the rules on the assumption that the range of the nuclear forces is infinite, and consists of Majorana, Wigner, Heisenberg, and Bartlett forces only, the Majorana and Wigner forces dominating.

The approximation of forces of infinite range is, of course, poor, but in the absence of anything else, these calculations may not be worthless especially as the calculations reported in § 3 show that the result continues to be true for finite range forces in some special cases.

We shall denote by (a) the wave-function of a single particle, magnetic quantum number a, in the shell nl_j; the wave-function of a hole, which is a Slater determinant of order $2j$, will be denoted by (a'). The wave-function of the state of spin I (with $M=I$) of two unlike particles (or particle and hole, etc.) is a sum of products of the functions (a) (or (a')) and will be denoted by

$$\psi(I)=\sum_{k=0}^{2j-I}\alpha_k\,\{(j-k)(I-j+k)\}$$

(or

$$\sum_{k=0}^{2j-I}\alpha_k\,\{(j-k)(I-j+k')\},\ \text{etc.})$$

(The α_k are Wigner coefficients; we do not require their exact form.)

$$\sum_k \alpha_k^2=1.$$

(The closed shells inside the nucleus can, of course, be neglected.)

We shall denote the energy integral

$$\oint (a)_1^*\,(b)_2^*\,P(c)_1\,(d)_2\,d^3r_1\,d^3r_2,$$

(where P is an exchange operator; W, M, H, or B, in the usual notation) by $(ab)\,(cd)_P$. We shall consider one case in detail, and quote the results for the others.

Class A(1). $j=l+\frac{1}{2}$, one particle of each sort in the shell. By direct calculation :

$$\left.\begin{aligned}
&(ab)\,(cd)_W=\delta_{ac}\,\delta_{bd},\\
&4j^2(ab)\,(cd)_B=\delta_{ac}\,\delta_{bd}\,(2j^2+2ab)\\
&\qquad+\delta_{a,\,c+1}\,\delta_{b,\,d-1}\,\{(j+a)(j-a+1)(j-b)(j+b+1)\}^{1/2}\\
&\qquad+\delta_{a,\,c-1}\,\delta_{b,\,d+1}\,\{(j-a)(j+a+1)(j+b)(j-b+1)\}^{1/2},\\
&(ab)\,(cd)_M=(ab)\,(dc)_B,\\
&(ab)\,(cd)_H=(ab)\,(dc)_W.
\end{aligned}\right\}\qquad(1)$$

I, the total spin of the nucleus, can take every value from 0 to $2j$, and since $(I_x+iI_y)\,\psi(I)=0$,

$$\{(2j-I-k)(I+k+1)\}^{1/2}\,\alpha_k+\{(k+1)(2j-k)\}^{1/2}\,\alpha_{k+1}=0\quad . \quad . \quad (2)$$

so that

$$\alpha_k=(-1)^{I+1}\,\alpha_{2j-I-k}.\quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (3)$$

Thus the binding energy $E_P(I)$, of the state of spin I due to the forces P is in arbitrary units

$$E_P(I)=\sum_{k=0}^{2j-I}\sum_{m=0}^{2j-I}\alpha_k\,\alpha_m\,(j-k\ I-j+k)\,(j-m\ I-j+m)_P.$$

Hence $E_W(I)=1$, from (1).

$$\begin{aligned}4j^2E_M(I)=&\sum_{k=0}^{2j-I}\alpha_k\,[\alpha_{2j-I-k}\,\{(2j-k)(I-k)+k\,(2j-I-k)\}\\&+\alpha_{2j-I-k+1}\,\{k(I+k)(2j-k+1)(2j-I+k+1)\}^{1/2}\\&+\alpha_{2j-I-k-1}\,\{(k+1)(I+k+1)(2j-k)(2j-I-k)\}^{1/2}]\\=&(-1)^{I+1}\,(I(I+1)-2j),\text{ from (2) and (3).}\end{aligned}$$

$$4j^2E_B(I)=(I(I+1)-2j),$$

$$E_H(I)=(-1)^{I+1}.$$

Since the Majorana forces dominate, the ground state is that for which $E_M(I)$ is the largest, i.e. $I=2j$, in accordance with (iii′).

Class $A(2)$. If, instead of two particles in the previous case, we have two holes, the results are exactly the same.

Class $A(3)$. If, however, we have one hole and one nucleon, we obtain :

$E_W(I)=-1$,
$E_B(I)=(I(I+1)-2j)/4j^2$,
$E_M(I)=0,\ I\neq 0,\ 1:\ =$a complicated expression $<0,\ I=0,\ 1$.
$E_H(I)=0,\ I\neq 0:\ =-2,\ I=0$.

In this case only the Bartlett forces separate the states of high spin, and these favour rule (iii′), provided the coefficient is positive, which is so for all proposed forms of the total P.

Class $A(4)$, (5), (6). If $j=l-\frac{1}{2}$, similar results hold.

In Class A, therefore, the rule is justified in the strict form for the infinite range case. Since we are concerned only with light nuclei, this is a not impossible approximation.

For *Class B* nuclei, the demonstration can be made for a general radial dependence of the nuclear forces. We shall suppose that it is the second particle which is in a state of spin $\frac{1}{2}$. Then

$$\psi(j+\tfrac{1}{2})=(j)_1\,(\tfrac{1}{2})_2,$$

$$\psi(j-\tfrac{1}{2})=[(j-1)_1\,(\tfrac{1}{2})_2-\sqrt{2j}(j)_1\,(-\tfrac{1}{2})_2]/(2j+1)^{1/2},$$

$$E_P(j+\tfrac{1}{2})=(j\tfrac{1}{2})\,(j\tfrac{1}{2})_P,$$

$$E_P(j-\tfrac{1}{2})=\frac{(j-1\ \tfrac{1}{2})(j-1\ \tfrac{1}{2})-2\sqrt{2j}(j-1\ \tfrac{1}{2})(j\ -\tfrac{1}{2})+2j(j\ -\tfrac{1}{2})(j\ -\tfrac{1}{2})}{(2j+1)}.$$

Now, imagining the indicated operations to be performed under the sign of integration, and remembering that the conjugate complex of the first two wave-functions is taken, we have :—

$$\begin{aligned}(j\ -\tfrac{1}{2})(j-1\ \tfrac{1}{2})&=(j\ -\tfrac{1}{2})\,[(I_x-iI_y)(j\ \tfrac{1}{2})_P-(j\ -\tfrac{1}{2})_P]/\sqrt{2j}\\&=\{[(I_x+iI_y)(j\ -\tfrac{1}{2})]\,(j\ \tfrac{1}{2})_P-(j\ -\tfrac{1}{2})(j\ -\tfrac{1}{2})_P\}/\sqrt{2j}\\&=\{(j\ \tfrac{1}{2})(j\ \tfrac{1}{2})_P-(j\ -\tfrac{1}{2})(j\ -\tfrac{1}{2})_P\}/\sqrt{2j}\end{aligned}$$

and similarly,

$$(j-1\ \tfrac{1}{2})(j-1\ \tfrac{1}{2})_P=[(2j-1)(j\ \tfrac{1}{2})(j\ \tfrac{1}{2})_P+(j\ -\tfrac{1}{2})(j\ -\tfrac{1}{2})_P]/2j$$

so that $\Delta E_P=E_P(j+\frac{1}{2})-E_P(j-\frac{1}{2})=\dfrac{2j+1}{2j}\,((j\ \tfrac{1}{2})(j\ \tfrac{1}{2})_P-(j\ -\tfrac{1}{2})(j\ -\tfrac{1}{2})_P)$.

Then, writing down the integrals,

$\varDelta E_W=\varDelta E_M=0$ in all cases ; and we have also :—

Class $B(1)$. $j=l+\frac{1}{2}$.

$$\varDelta E_B=\frac{2j+1}{2j}\oint |Y_l^l(\theta_1)|^2 |R_{nl}(r_1)|^2 |R_{n'\,s}(r_2)|^2\, V(r_{12})\, d^3r_1\, d^3r_2,$$

$$\varDelta E_H=\frac{2j+1}{2j}\oint Y_l^{l*}(\theta_1)\, Y_l^l(\theta_2)\, R_{nl}(r_1)\, R_{nl}(r_2)\, R_{n'\,s}(r_1)\, R_{n'\,s}(r_2) \times V\,(r_{12})\, d^3r_1\, d^3r_2.$$

In the integral for $\varDelta E_B$, the integrand is positive. Hence $\varDelta E_B>0$. Also

$$\varDelta E_B\pm\varDelta E_H=\frac{2j+1}{4j}\oint |R_{nl}(r_1)\, Y_l^l(\theta_1)\, R_{n'\,s}(r_2)\pm R_{nl}(r_2)\, Y_l^l(\theta_2)^*\, R_{n'\,s}(r_1)|^2 \times V\,(r_{12})\, d^3r_1\, d^3r_2$$

which is positive for the same reason. Hence $E_B>|E_H|$. The resulting E_P is thus certainly positive of P has either the form proposed by Rosenfeld (1948) ($=0{\cdot}93M-0{\cdot}13W+0{\cdot}46B-0{\cdot}26H$) or that proposed by Serber ($P=0{\cdot}4M+0{\cdot}4W+0{\cdot}1B+0{\cdot}1H$). Thus the ground state certainly has $I=j+\frac{1}{2}$ in accordance with the rule.

This is the simplest case : we shall not write out the integrals for the other cases, but express the results in words.

Class $B(2)$. $j=l-\frac{1}{2}$.

By an argument more complex than that above, but still elementary, we can now show that $\varDelta E_B<0$. Also $-\varDelta E_B>|\varDelta E_H|$*, so that, as before, we have $I=j-\frac{1}{2}$, in accordance with the rules, for all forms of V and P.

It is to be noted that the proof is possible in case B, only because of the failure of the dominant Majorana and Wigner forces to separate the two states, so that spin-dependent forces, as in the deuteron, favour parallel spins.

§ 3. EFFECTS OF FINITE RANGE OF NUCLEAR FORCES

For forces of finite range, a general calculation in case A is no longer possible. Calculations have been carried out for the nuclei ^{18}F and ^{22}F on the (false) assumption that these nuclei have orbitals in the $d_{5/2}$ shell only. The technique is described in a following paper (Hitchcock 1954). It is found that, except for very short range forces (iii′) continues to hold, but that for these the calculated spin of ^{18}F is 0, and of ^{22}F, 4. Similar calculations on Sc^{42} and Sc^{48} together with those of Kurath (1951), and those of Flowers and Edmonds (1952) suggest the following rule :—

For cases $A(1)$, (2), (4) and (5), (iii′) holds, except for very short range forces, or large nuclear radii ; for cases $A(3)$ and (6) rule (iii′) holds for long range forces (or small nuclear radii), but for forces of shorter range (or larger nuclear radii) the calculated spin is $(2j-1)$, so that (iii) holds. No general proof of this proposition, however, is available.

---

* This inequality cannot be proved with the same generality as the others, but is certainly true for all the usually assumed forms of V.

For forces of sufficiently short range, Pryce (1952) has shown that, in shells of large j, the least possible spin is probably always preferred. All the calculations mentioned above, however, indicate that this ceases to be true at the range of forces which is of interest, at any rate in the light nuclei considered here.

An attempt to extend Class B to include nuclei containing a $p_{1/2}$ nucleon fails, since we no longer have $\Delta E_M = 0$. A similar proof to those of class B does, however, readily apply to the two extreme cases of infinite range and zero range forces, since for these we do have $\Delta E_M = 0$.

We conclude that the agreement between the proposed empirical rules for the spins of odd–odd nuclei and experiment is partially accounted for. We have already pointed out (§ 1) that the spin-dependent forces must always favour the truth of rules (ii) and (iii′). If there is indeed a tendency for the rules to be obeyed in other classes of nuclei, this may suffice to account for it. Certain attempts to extend these general calculations have proved unsuccessful, and more detailed calculations (Hitchcock 1954) indicate that the rules no longer hold theoretically.

The author's thanks are due to Professor M. G. Mayer, and to Mr. J. M. C. Scott, for valuable criticism and discussion.

References

Edmonds, A. R., and Flowers, B. H., 1952, *Proc. Roy. Soc.* A, **214** 515; **215,** 120.

Hitchcock, A., 1952, *Proc. Phys. Soc.* A, **65,** 661; 1954, *Phil. Mag.*, **45,** 385.

Kurath, D., 1951, *Ph.D. Thesis,* University of Chicago.

Nordheim, L. W., 1951, *Rev. Mod. Phys.*, **23,** 322.

Nussbaum, R. H., van Lieshout, R., and Wapstra, A. H., 1953, *Physica,* **19,** 135.

Pryce, M. L. H., 1952, *Proc. Phys. Soc.* A, **65,** 733.

Rosenfeld, L., 1948, *Nuclear Forces* (Amsterdam: North Holland), p. 234.

XLVI. *Spins of Odd–Odd Nuclei in the j–j Coupling Model : II*

By A. Hitchcock*
Cavendish Laboratory, Cambridge†

[Revised MS. received December 14, 1953]

Abstract

The spins of a number of odd odd nuclei are calculated on the basis of the spin–orbit coupling model. The study is limited to light and medium nuclei for which experimental data are available. The agreement between theory and experiment is generally good. No systematic rules are proposed, and it appears that no simple ones exist, except in the cases previously treated.

§ 1. Introduction

It has already been shown (Hitchcock 1954) hereafter referred to as I, that the empirical rules proposed by Nordheim (1951) for the spins of odd–odd nuclei can be justified in certain restricted cases. It is the object of this paper to extend the investigation of such spins to other nuclei, in order to see if the rules may be more generally applicable. (The rules are quoted in I, and will hereafter be referred to as (i), (ii), (iii) and (iii′).)

The *j–j* coupling model is taken as a basis for all the work, and, except in one case, it is assumed that nuclear forces are entirely two-body in character, though different exchange operators are considered. We also assume charge independence of nuclear forces, that is, that proton–proton, proton–neutron, and neutron–neutron forces all have the same form ; and neglect the Coulomb force entirely. (It has been found by trial in a few cases including ^{50}V, one of the heavier nuclei studied here, that its inclusion does not affect the results provided that no variation in the core configuration is allowed.)

We shall refer to the closed subshells of a nucleus as the *core*, and the remaining nucleons will be called *orbital* nucleons. In all calculations of this type we are concerned with the lowest excited states only. Configurational mixing, both in the core and in the orbital nucleons is neglected and the core itself can therefore be neglected, except in so far as it provides the source of the fictitious potential well in which the orbital nucleons move : for on account of the complete spherical symmetry of closed shells, the energies of orbital nucleons in the field of the core are equal, provided no orbital nucleon is excited into another shell. Thus we need only consider the interactions between the orbital nucleons.

---

* Communicated by the Author.

† Now at Department of Atomic Energy I. G., Risley, Warrington, Lancs.

From this, it follows without difficulty, that the low-energy level schemes of what we here call conjugate nuclei are identical, apart from small effects due to differences in nuclear radius. The nuclei conjugate to a nucleus with N_1 protons in the $n_1l_1j_1$ shell and N_2 neutrons in the $n_1l_1j_1$ shell are defined to be the nuclei with $(2j_1+l- N_1)$ protons (or neutrons) in the $n_1l_1j_1$ shell and $(2j_2+l-N_2)$ neutrons (or protons) in the $n_2l_2j_2$ shell.

In many cases, the level schemes of conjugate nuclei are dissimilar. This must be ascribed in part to the effects of different nuclear radius, or to the Coulomb force, but these are too small to account fully for the effects, which are therefore due to partial failure of the j–j coupling model itself. (We shall say that the model ‘ fails ’ if the effects of configurational mixing become large enough to affect the results.)

In general, indeed, these calculations, and the comparison of their predictions with experiment, provide us with a sensitive test of the extent of the validity of the assumption of spin–orbit coupling. In an odd–odd nucleus, which has none of the simple symmetry properties of other kinds, there are many possible ground states, between which we cannot choose without explicit calculation. It is thus satisfactory that only one definite disagreement with experiment is found.

§ 2. FORCES AND RADIAL WAVE-FUNCTIONS

The greatest uncertainty in all calculations of this kind arises from our lack of knowledge of the form of the two-body forces between nucleons. For want of better information we have assumed that the inter-nucleon potential is of one of the forms :

(i) $V(r)=V_0\exp(-\alpha r^2)P$, referred to as G, for Gaussian, in the succeeding, where

$$V_0=42 \text{ MeV},$$
$$\alpha=0\cdot1089\times10^{26}\text{ cm}^{-2}.$$

(ii) $V(r)=V_0\,(\exp(-\alpha r)/r)P$, referred to as Y, for Yukawa, in all the succeeding, where

$$V_0=89\times10^{-13}\text{ MeV cm},$$
$$\alpha=0\cdot858\times10^{13}\text{ cm}^{-1}.$$

(iii) $V(r)=V_0\delta(r)P$, referred to as δ in all the succeeding.

The numerical values are taken from those derived by Blatt and Jackson (1950). P is supposed to be an exchange operator : we take

(*a*) $P=0\cdot8M+0\cdot2H$ referred to as MH, or

(*b*) $P=0\cdot4M+0\cdot4W+0\cdot1B+0\cdot1H$, referred to as S, for Serber, in the succeeding.

The two forms are indistinguishable in case δ.

The tensor force has been neglected. In a large nucleus, where its range is comparatively small, this approximation may be justifiable, but as the calculation on ^{6}Li (*q.v.*) shows, it would be desirable to consider it. It is hoped to do this in future work.

Further uncertainty arises in the form to be adopted for the radial wave-functions. In theory, a sufficiently accurate approximation to these may be obtained by a variational calculation, but in practice it is also necessary that they be in a form suitable for the performance of the integrations arising. We have therefore chosen in case G

$R_{nl}(r)=\exp(-\beta r^2)r^lH_{nl}(\beta r^2)$—‘ harmonic oscillator ’ functions and in case Y

$R_{nl}(r)=\exp(-\beta r)r^lL_{nl}(\beta r)$—‘ hydrogen ’ functions, where in each case the H and L are the suitable polynomials.

β is determined, following Kurath (1951), by fitting the wave-function as well as possible to the corresponding wave-function of a square well of radius $r_0=1{\cdot}48\times10^{-13}A^{1/3}$ cm. In case δ the form chosen is irrelevant, provided configurations with nucleons in varying shells are not considered, as we shall not. Since this method is uncertain, a range of values of β has been used in all the calculations, and it is found that in most cases the order of levels is not sensitive to the value chosen. However, the actual level separations depend very sensitively on β, so that it is not possible to predict the values of the separations to an accuracy sufficient to make comparison with experiment worth while. Where the calculated spins differ for larger and smaller admissible values of the nuclear radius, we shall distinguish the case of the larger radius by a suffix 1, and the smaller by a suffix 2, thus : G_1, YMH_2. If one of these is less likely than the other it will be bracketed.

We may note that the wave-functions of case Y are probably less like the actual ones than those of case G, and the use of the former, when two or more shells are involved, leads to violent anomalies (Hitchcock 1953). Case G is therefore to be preferred if the results differ. In fact, as we shall see, case GS nearly always agrees with experiment.

§ 3. CLASSIFICATION OF MANY-PARTICLE WAVE-FUNCTIONS

It may happen, and in general does, that a number of orbital nucleons can have more than one configuration of the same spin and isotopic spin. A further classifying observable is, therefore, required. Ideally it is desirable to find a third observable which is actually conserved under the inter-nucleon forces, and which, like spin and isotopic spin, lends itself readily to group-theoretical treatment ; but no such observable seems to exist. It is necessary therefore to consider a linear combination of the wave-functions arising in the classification chosen, calculate the energy, and minimize. A classification which approximately satisfies the ideal condition reveals itself by small cross-terms in the parametric expression for the energy.

Flowers (1952) has introduced the concept of seniority, which satisfies the required conditions almost exactly. This is certainly the most elegant, and probably the best classification proposed. Kurath (1951) obtained wave-functions which contained a minimal number of terms, subject to being eigenfunctions of spin and isotopic spin. For the simple cases he treated this is adequate, but the method is of little use in more complicated cases.

As the work of Flowers was unpublished at the time these calculations were begun it was decided to adopt a classification in which proton spin and neutron spin were separately conserved. This has the following advantages :

(*a*) It also approximately satisfies the ideal condition.

(*b*) It lends itself to physical interpretation, and enables us to see at once to what extent rule (i) is valid.

(*c*) It breaks a complicated calculation into a number of small sections, making checking easier, and reducing the likelihood of computational error.

(*d*) It is found by trial that states of very large proton and neutron spins occur to a negligible extent in the low excited states. This enables the neglect of these states, and consequent great simplification of the calculations.

(*e*) It should be noted however that the states obtained are not eigenstates of the isotopic spin.

§ 4. SPECIFIC NUCLEI

We shall consider conjugate nuclei together, where both exist, and summarize the results only, without entering into the details of the calculations, or quoting numerical results. We shall not treat nuclei in which one nucleon is in an $s_{1/2}$ state, for which a general proof has been given in I, unless some special consideration makes it desirable. The quoted experimental results are derived from Way *et al.* (1950).

^{6}Li *and* ^{10}B

Measured spin of ^{6}Li	1
Measured spin of ^{10}B	3
Calculated spin of ^{6}Li	1
Calculated spin of ^{10}B	3

The work on ^{6}Li has already been reported (Hitchcock 1952 a). In this case alone the tensor force was considered. It is shown that it is possible to account for the spin 1 by assuming strict *j–j* coupling, although the contrary has been asserted. The effect of the tensor force, however, decreases rapidly with increase in nuclear radius, and a rough calculation for ^{10}B indicates that in this nucleus its effect is not sufficient to invert the levels of spins 3 and 1. The position with regard to excited states is equally satisfactory. ^{6}Li has two excited states of spins 3 and 0, which occur in the order predicted. The two lowest excited states of ^{10}B have spins 1 and 0 according to both theory and experiment.

It should be pointed out that in spite of these satisfactory results, it is doubtful if there is, in fact, true *j–j* coupling in ^{6}Li.

Much work has been done on these nuclei assuming intermediate coupling but neglecting the tensor force (Inglis 1953, Racah and Zeldes 1951, Zeldes 1953). It seems probable that the effects of both intermediate coupling and of the tensor forces should properly be taken into account. The work of Zeldes seems to suggest that sufficiently large level separations cannot be obtained on the basis of intermediate coupling alone. (Inglis' calculation assigns the nuclear parameters to fit the level schemes in a manner which seems to allow different values of these parameters in the two nuclei.)

^{14}N

Measured spin	1
Calculated spin	1

This nucleus was considered, allowing for the possibility of $p_{1/2}-p_{3/2}$ mixing, in order to see if the remarkably long β-half-life of ^{14}C, and the associated lack of the ground-state transition in ^{14}O, could be accounted for by ' accidental ' vanishing of the nuclear matrix element. A tensor force, of the same range as the ordinary forces, was assumed. In fact too much mixing is found, and the *ft* value is increased in consequence, assuming Gamow–Teller selection rules, by a factor 3 only.

The predicted magnetic moment is 0·63. If $p_{3/2}$ mixing is neglected we get 0·37. The measured value is 0·40. This suggests that if the mixing were less for some reason (e.g. if the $p_{3/2}-p_{1/2}$ separation is greater in ^{14}N than in ^{7}Li, the value chosen) there would be better agreement. But in fact no improvement is found if the amount of mixing is adjusted to fit the magnetic moment, though consideration of the mixing of higher states could improve the fit.

^{18}F

Experimental spin	1 or 0*
Calculated spin, Y and G_2	1
Calculated spin, G_1 and δ	0

Here the possibility of $1d_{5/2}$–$2s_{1/2}$ mixing was considered. As already reported (Hitchcock 1953) the pairing energy of the $2s_{1/2}$ nucleons is greater than that of the $1d_{5/2}$ nucleons, for cases of long range forces. Consequently, the ground and first excited (calculated spin 0) states are predominantly $(s_{1/2})^2$ configurations, the lowest $(d_{5/2})^2$ configurations coming much higher. This may account for the lack of low excited states in this nucleus (Hitchcock 1953).

^{22}Na

Measured spin	3
Calculated spin, δ, G and (YS_1)	2
Calculated spin, YMH	5
Calculated spin, YS_2	3

---

* In view of the 0→0 transition in ^{14}O, a not dissimilar nucleus, the value 0 cannot be excluded.

Agreement with experiment is unsatisfactory. $2s_{1/2}$ mixing has been neglected, but a rough calculation indicates that its inclusion is unlikely to improve the situation. The β-transition to an excited state of ^{22}Ne has a log *ft* value of 7. Since the shell model does not allow for parity change here, this is exceptionally large. The calculations do not uphold the suggestion that this is due to the re-arrangement of the configuration $(d_{5/2})^3_{j=3/2}$, as rule (i) suggests, since 30% of the wave-function corresponds to the configuration $(d_{5/2})^3_{j=5/2}(d_{5/2})^3_{j=5/2}$.

It is possible, however, though unlikely, that $s_{1/2}$ mixing occurs to such an extent that not only is the spin 3 state depressed to the ground state but that the β-transition is '*l*-forbidden' (*vide* ^{32}P). This solution is advanced with little conviction.

^{24}Na *and* ^{20}F

Measured spin of ^{24}Na	4
Experimental spin of ^{20}F	2 or 3 (??)
Calculated spin, *GMH*, *YS*, YMH_1	5
Calculated spin, δ, *GS*, (YMH_2)	4

$2s_{1/2}$ mixing has again been neglected. The experimental indications for ^{20}F are conflicting, so that disagreement with experiment is not established here. For ^{24}Na agreement is excellent. The level schemes of these nuclei have been extensively investigated. There is no evidence of the correspondence expected in conjugate nuclei.

$2s_{1/2}$ *mixture in* ^{20}F, ^{22}Na, *and* ^{24}Na

The results for ^{18}F suggest that the approximation of neglect of $2s_{1/2}$ mixture in the preceeding nuclei may be very poor. It is of interest to proceed to the opposite extreme, and suppose—as indeed the calculation for ^{18}F suggests—that there is complete replacement of $d_{5/2}$ pairs by $s_{1/2}$ pairs in these cases. Calculated results now become :

^{20}F, Configuration $((s_{1/2})^2d_{5/2})$, $d_{5/2}$.

Calculated spin, δ	0
Calculated spin, *G* and *Y*	5

Configuration $((s_{1/2})^2d_{5/2})$, $s_{1/2}$.

Calculated spin	3 (see I).

^{22}Na, Configuration $d_{5/2}$, $d_{5/2}$.

As ^{20}F, first above.

^{24}Na : The new configuration $(d_{5/2})^3(d_{5/2})$ is the conjugate of the former $(d_{5/2})^5(d_{5/2})^3$, so that the results are unaltered. In this case, therefore, intermediate mixture will also probably lead to the same result.

There is little improvement ; and in any case, the spin 3/2 of ^{21}Ne shows that its configuration is not $((s_{1/2})^2\,d_{5/2})$, $(s_{1/2})^2$ as the assumption above would require.

^{32}P

$1d_{5/2}$–$2s_{1/2}$ mixing was considered. The log *ft* value of the β-transition to the closed-shell nucleus ^{32}S is 7·9, and the Kurie plot is straight. The shell assignments do not allow of parity change, so this is presumably an allowed transition. Nordheim (1951) classifies it as 'l-forbidden'; suggesting, in effect, that transitions with $\Delta I=1$, $\Delta l=2$ ($s_{1/2}-d_{3/2}$, etc.) cannot be placed in the same class as other allowed transitions ; and indeed the Gamov–Teller allowed matrix element vanishes for such a transition. The observed decay must, therefore, be due to a $d_{5/2}$ contamination in ^{32}P, which must be of the order 0·1% to account for the observed half-life. For a suitable and admissible choice of the nuclear radius this can be done in both cases G and Y, and at the same time a separation between ground and first excited states of the right (70 kev) order of magnitude obtained.

It is pertinent to ask why a precisely similar state of affairs does not apply in ^{34}P, which has a log ft value of 5. The answer lies in the fact that the interaction between the $(d_{3/2})^{-1}$ neutrons and the $(d_{5/2})^{-1}$ protons in the ground (spin 1) state of ^{34}P is much greater than that between the single $d_{3/2}$ neutron and the $(d_{5/2})^{-1}$ protons in ^{32}P, whereas the interactions of the two neutron configurations with an $s_{1/2}$ proton are the same. Thus the $d_{5/2}$ contamination in ^{34}P is appreciably greater than in ^{32}P, and the ft value of the β-transition correspondingly less. Thus the *ad hoc* assignment by Nordheim of the fifteenth proton to an $s_{1/2}$ state in ^{32}P and to a $d_{5/2}$ state in ^{34}P has approximate justification.

Another explanation is possible. If the spin of ^{32}P is in fact 2, as is indicated theoretically as a possibility, then the proton configuration is mainly $(d_{5/2})^{-1}$ and we have a second-forbidden transition with $\Delta I=2$, $\Delta l=0$. Following Nordheim's nomenclature we may call such transitions 'l-preferred', since calculation of the matrix element indicates that the ft values of such transitions should be reduced by a factor of the order 3×10^{-3}, which leads us to expect log ft values between 7·5 and 8·5 for such transitions. This fits the value for ^{32}P, but the allowed shape of the β-spectrum makes this explanation less plausible than the previous one.

^{36}Cl

Measured spin	2 (3 ?)
Calculated spin, GS, δ	2
Calculated spin, Y, GMH	3

Agreement is satisfactory.

^{38}Cl

Experimental spin	0, 1, or 2
Calculated spin, δ	2

The shell model allows no spin less than 2, so it is satisfactory that 2 is found. It is the most tightly bound by so much that it is likely to be so in cases G and Y also for all reasonable values of the nuclear radius. The first excited state is predicted to have spin 5, which suggests isomerism, but a finite range calculation, will be required to establish if the model really predicts this.

^{40}K

Measured spin	4
Calculated spin, δ	4

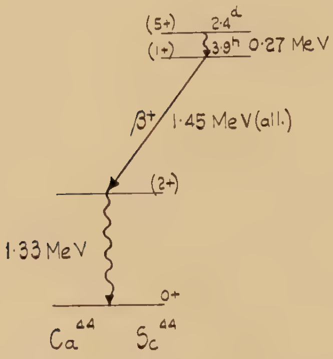
According to Kurath (1952) the calculated spin is 4 in case *G* also.

^{44}Sc and ^{52}Mn

The calculations (case δ only) indicate that the spins of the low excited states, in ascending order, are 1 (ground), 6, 4, 5.*

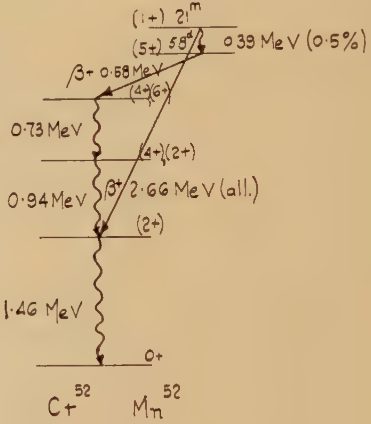
These nuclei both show isomerism and, according to Goldhaber and Sunyar (1952), the isomeric transitions are M4 in ^{52}Mn, and M4 or E4 in ^{44}Sc ; an E4 transition implies parity change, which is not acceptable to the shell model. M4 is therefore assigned to both transitions. This is slight confirmation of the theory of conjugate nuclei.

Fig. 1



Decay scheme of ^{44}Sc . Assigned values of spin and parity given in brackets ; (all.) indicates allowed transition.

Fig. 2



Decay scheme of ^{52}Mn.

That the ground state is predicted to be of low spin is satisfactory, at first sight, and we must assign spin 5 to the second isomer. The depression of the state of spin 5 below the (very close) states of spin 4 and 6 can very probably be due to the effect of finite range nuclear forces—a similar depression is predicted in ^{46}Sc (*q.v.*). We therefore assign spin 1 to 4-hour ^{44}Sc and 21-minute ^{52}Mn* ; and assign spin 5 to $2\frac{1}{2}$-day ^{44}Sc* and 6-day ^{52}Mn. It is difficult to see, if these assignments are correct, why the β-transition to the ground state of the daughter nucleus is not observed, but the lowest state of spin 2 is predicted to be the seventh excited state, and cannot be depressed to the ground state by considerations of finite range. Agreement with experiment is thus not satisfactory.

---

* Dr. Flowers informs me that there is an error in the calculations reported in his paper (1951) which disagrees with those reported here.

^{46}Sc and ^{50}V

Measured spin of ^{50}V	6
Experimental spin of ^{46}Sc	4 or 5
Calculated spin, δ	6
Calculated spin, G and Y	5

The work of ^{50}V has been reported previously (Hitchcock 1952 b), and the same calculation applies for ^{46}Sc. It is certain, that for all nuclear radii in the relevant interval the calculated spin will be either 6 or 5, and it may be that the slightly larger nuclear radius of ^{50}V is adequate to ensure its conformity with case δ, whereas it is necessary to invoke finite range forces to explain the spin of ^{46}Sc. The difference in radius must be small, however, and the calculations do not indicate that the cross-over point is in the region of interest.

^{46}Sc shows E3 isomerism (Goldhaber and Sunyar 1951). The calculations suggest spin 2 for the isomer, which is in accordance with the decay scheme. No such isomerism, however, is apparent in ^{50}V.

§ 5. CONCLUSIONS

We conclude that the assumption of strict *j–j* coupling is not inconsistent with the observed spins of light and medium nuclei, especially, it will be noted, in case *GS*. There is no reason to suppose that it will not also account for those of heavy nuclei, but there are few experimental data for comparison, and shell assignments are uncertain, so the laborious calculations are not profitable.

Agreement with experiment is generally satisfactory (except for ^{22}Na, and to a lesser extent for ^{44}Sc and ^{52}Mn) so far as spins are concerned. The agreement of the level schemes of conjugate nuclei is not good, except for the $\Delta I=4$ isomeric transitions in ^{44}Sc and ^{52}Mn. A tendency for the spins to increase through a shell, pointed out by King and Peaslee (1953) is, of course, contrary to the *j–j* coupling model, and could not be justified by these calculations.

There is no theoretical justification of rules (ii), (iii) or (iii′). except in the cases considered in I.

REFERENCES

BLATT, J. M., and JACKSON, J. D., 1950 a, *Rev. Mod. Phys.*, **22,** 77; 1950 b, *Phys. Rev.*, **76,** 18.
FLOWERS, B. H., 1951, *Proc. Roy. Soc.* A, **210,** 497.
GOLDHABER, M., and SUNYAR, A. W., 1951, *Phys. Rev.*, **83,** 906.
HITCHCOCK, A., 1952 a, *Proc. Phys. Soc.* A, **65,** 661; 1952 b, *Phys. Rev.*, **87,** 664; 1953, *Phil. Mag.*, **44,** 766; 1954, *Ibid.*, **45,** 379.
INGLIS, D. G., 1953, *Rev. Mod. Phys.*, **25,** 287.
KING, R. W., and PEASLEE, D. C., 1953, *Phys. Rev.*, **90,** 1001.
KURATH, D., 1951, *Ph. D. Thesis*, University of Chicago; 1952, private communication.
NORDHEIM, L. W., 1951, *Rev. Mod. Phys.*, **23,** 322.
RACAH, G., and ZELDES, N., 1951, *Phys. Rev.*, **79,** 1012.
WAY, K., FANO, L., SCOTT, M. R., and THEW, K., 1950, *U.S. Nat. Bur. Stand.*, Circ. 499 and supplements.
ZELDES, N., 1953, *Phys. Rev.*, **90,** 416.

XLVII. *Theory of the Effect of an Electric Field on Liquid Viscosity*

By R. EISENSCHITZ and G. H. A. COLE*
Queen Mary College, University of London†

[Received January 18, 1954]

ABSTRACT

If an insulating liquid is placed between the plates of a condenser the electric field gives rise to induced intermolecular forces which are proportional to the square of the field strength. On account of these forces there is a change in the radial distribution function. The field effect on the distribution function is calculated for the case of thermal equilibrium and for steady flow. The corresponding increment in viscosity compares not unfavourably with experimental facts.

§1. INTRODUCTION

THERE is, so far, no molecular theory of the effect of an electric field on the viscosity of liquids as observed by Andrade and Dodd (1950). In this paper an interpretation of this effect is attempted by considering the intermolecular forces which are induced by an electric field and their contribution to the flow of momentum. Ignoring some finer aspects a simplified molecular model is used by assuming that the molecules interact with central forces on which the forces of a molecular dipole may be superposed. The interaction of the molecules in an electric field is then obtained without any further assumption. The contribution of the induced forces to the coefficient of viscosity is arrived at by applying the theory of transport processes as developed by Kirkwood (1946) and Eisenschitz (1949).

§2. INTERACTION OF MOLECULES IN AN ELECTRIC FIELD

The average electric moment of a molecule is equal to

$$M=[\alpha+(\mu^2/3kT)]F=\alpha F+M_D \quad . \quad . \quad . \quad . \quad . \quad . \quad (1)$$

(Debye 1912) where α and μ are the electronic polarizability and the permanent moment ; $\mathbf{F}$ is the field vector.

Molecules in an electric field should accordingly interact like parallel dipoles with a potential energy of interaction

$$\chi_M=M^2K(\mathbf{r})+\ldots \quad . \quad . \quad . \quad . \quad . \quad . \quad (2a)$$

where $$K(\mathbf{r})=K(r,\,\vartheta)=(1-3\cos^2\vartheta)r^{-3} \quad . \quad . \quad . \quad . \quad . \quad . \quad (3)$$

$\mathbf{r}$ being the distance vector.

---

* Now at the Admiralty Research Laboratory, Teddington.
† Communicated by the Authors.

Ignoring at first the first term in (1) the validity of eqn. (2 *a*) is readily established. The potential energy of a pair of polar molecules in an electric field is equal to

$$\chi_F=\chi_{00}(r)+[r^2(\boldsymbol{\mu}_1\cdot\boldsymbol{\mu}_2)-3(\boldsymbol{\mu}_1\cdot\mathbf{r})(\boldsymbol{\mu}_2\cdot\mathbf{r})]r^{-5}-(\boldsymbol{\mu}_1+\boldsymbol{\mu}_2)\cdot\mathbf{F} \quad . \quad (4)$$

where $\boldsymbol{\mu}_1$, $\boldsymbol{\mu}_2$ $(\mu_1=\mu_2=\mu)$ are the molecular dipoles ; χ_{00} is the familiar potential energy of molecular interaction. An exact knowledge of the potential energy at small distances—where eqn. (4) does not apply—will not be required in this paper.

The force of interaction is $-\text{grad}\,\chi_M$ where

$$\chi_M=-kT\log\left[(1/16\pi^2)\int\exp(-\chi_F/kT)\,d(\boldsymbol{\mu}_1/\mu)\,d(\boldsymbol{\mu}_2/\mu)\right] \quad . \quad . \quad (5)$$

By expanding to the third power of the exponent it is found that

$$\chi_M=\chi_{00}-(\mu^4/3kTr^6)-(\mu^2F^2/3kT)+M_D{}^2K(r,\vartheta)$$

which is in accordance with (2 *a*).

The interaction of non-polar molecules is calculated by applying quantum mechanics to the electrons. The second order perturbation of energy is familiar and equal to $-\alpha F^2+\chi_{00}$. In the third order an expression is found which is very nearly equal to $(\alpha F)^2K(r,\vartheta)$ in accordance with eqn. (2 *a*).

The complete interaction of polar molecules is—for the simplified molecular model—obtained by successive application of perturbation theory to the movement of electrons and evaluation of the angular partition function of the molecular dipoles. The result is in agreement with (2 *a*) and conveniently written in the form

$$\chi_M=\chi_0-\alpha'F^2+M^2K(r,\vartheta) \quad . \quad . \quad . \quad . \quad . \quad (2\,b)$$

where χ_0 is the potential energy in absence of a field and α' is the mean polarizability.

In this paper eqn. (2 *b*) is applied to a pair of molecules in the interior of a liquid. F is then the internal field strength and related to the external field strength in terms of the dielectric constant ϵ according to

$$F=(1/3)(\epsilon+2)F_e. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (6)$$

§3. LIQUID IN THERMAL EQUILIBRIUM

If $f_0(r)$, $f_M(\mathbf{r})$ are the distribution functions in relative coordinates in absence or presence of an electric field, ‘ effective ’ potential energies of interaction Φ_0, Φ_M are defined by writing

$$f=\exp(-\Phi/kT)$$

and the negative gradient of the effective potential energy may be regarded as the force determining the relative movement of the molecules. The effect of an electric field on the properties of a liquid is determined by its effect on Φ_0.

A rough estimate of this effect is made by assuming that the difference of Φ_M and $\chi_M+\Phi_0$ arises from the interaction of a shell of parallel dipoles with a molecule when another molecule is located at the centre of the shell. Let the radial distribution of the dipoles in the shell be specified in terms of a distribution function $f_S(r)$ then the effective potential is found to be

$$\Phi_M=\Phi_0+\chi_M+8\pi M^2(\partial^2/\partial z^2)\left\{r^{-1}\int_0^r f_S(r')r'^2\,dr'+\int_r^\infty f_S(r')r'\,dr'\right\}. \quad . \quad (7)$$

Using a distribution function f_S of simple analytical form and corresponding to 10 nearest neighbours, expression (7) was evaluated. The result was applied in numerical tests, as specified in §6, but not for calculating quantities to be compared with experiment.

Improving on this calculation, consider the energy of interaction of the molecules labelled as i and j

$$\chi_M(i, j)=\chi_M(\mathbf{r}_{ij})=\chi_0(r_{ij})+M^2K_{ij}$$

where K_{ij} is written for $K(\mathbf{r}_{ij})$. The probability distribution for the simultaneous positions of the molecules 1 and 2 is given by the expression

$$f_M(1, 2)=(V/Z)\int \exp\left(-(\tfrac{1}{2}kT)\sum_{i,j}\chi_M(i, j)\right) d\mathbf{r}_{23}\ldots d\mathbf{r}_{2N}$$

where V is the volume, $Z(T, V, M)$ is a normalization factor satisfying the relation $[M^{-1}(\partial Z/\partial M)]_{M=0}=0$; in the integrand molecule 2 is placed at the origin of the coordinates.

Expanding f_M in powers of M^2, the coefficient of M^2 is equal to the required increment of the distribution function. It is equal to

$$kT(\phi_M-\phi_0)f_0(r_{21})=K_{21}f_0(r_{21})+2N\int f_0^{(3)}(1, 2, 3)K_{23}\,d\mathbf{r}_{23}$$
$$+N^2\int f_0^{(4)}(1, 2, 3, 4)K_{34}\,d\mathbf{r}_{23}\,d\mathbf{r}_{24}. \quad . \quad . \quad . \quad (8)$$

Here $f_0^{(3)}$ and $f_0^{(4)}$ are the distribution functions for triplets and quadruplets of molecules and N is the number of molecules present.

Equation (8) does not depend on any approximation other than implied in the use of a constant internal field. The right-hand side of (8) can, however, be evaluated only if the unknown distributions are eliminated conveniently by means of the 'superposition approximation'

$$\left.\begin{aligned} f_0^{(3)}(1, 2, 3)&=f_0(1, 2)f_0(1, 3)f_0(2, 3),\\ f_0^{(4)}(1, 2, 3, 4)&=f_0^{(3)}(1, 2, 3)f_0(1, 4)f_0(2, 4)f_0(3, 4).\end{aligned}\right\} \quad . \quad . \quad (9)$$

By entering with (9) into (8), Φ_M is expressed in terms of known distribution functions.

The last term in (8) is, in spite of the factor N^2, much smaller than the preceding term on account of the angular function in the integrand which reduces the integral to zero unless the four molecules are situated close to each other. It was accordingly possible to neglect this term. The remaining terms are reduced to the following expression

$$\Phi_M=\Phi_0-2M^2P_2(\cos\vartheta)[r^{-3}+2\pi NJ(r)] \quad . \quad . \quad . \quad . \quad (10)$$

where $$J(r)=\int_0^\infty\int_0^\pi f_0(r'')f_0(r')r'^{-1}\sin\theta\,d\theta\,dr'$$

and $$r''=(r^2+r'^2-2rr'\cos\theta)^{1/2}$$

and P_2 is the Legendre polynomial. Expression (10) may be regarded as a better approximation than (7) but its evaluation requires extensive numerical work. Expressions (7) and (10) can be written in a common form in terms of two functions of r, denoted by Φ_1 and Φ_2

$$\Phi_M=\Phi_0+M^2[\Phi_1+2\Phi_2P_2(\cos\vartheta)] \quad . \quad . \quad . \quad . \quad . \quad (11)$$

which will be used in the following calculations.

§4. THE STEADY NON-UNIFORM STATE

The distribution $f_A(r)$ which is appropriate to a liquid in steady flow, is determined by the Smoluchowski equation

$$\partial f_A/\partial t=\mathrm{div}\{(kT/\gamma m)\,\mathrm{grad}\,f_A+[(\mathrm{grad}\,\phi_M/\gamma m)-\mathbf{c}]f_A\}=0. \quad . \quad (12)$$

Here $\mathbf{c}(\mathbf{r})$ is the macroscopic velocity, m is the molecular mass and γ a friction constant. Let

$$c_x=c_z=0 \qquad c_y=Az$$

where $A/2$ is the constant yz-component of the rate of shear. Neglecting terms of the order A^2, eqn. (12) can be solved in terms of a function $W_M(\mathbf{r}): f_A=f_M[1+AW_M]$. Expanding W_M in associated Legendre polynomials only one term in the expansion makes a contribution to viscosity :

$$W_M(r,\vartheta,\phi)=u_M(r)P_2^{\,1}(\cos\vartheta)\sin\phi+O(M^4). \quad . \quad . \quad (13)$$

In accordance with previous work (Eisenschitz 1949) the function u_M has to satisfy the condition

$$r^3u_M(r)=0 \quad \text{for} \quad r\to\infty. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (14)$$

At small values of r ($r<B\div 3\times 10^{-9}$ cm) u_M may be replaced by zero without a serious error being made.

Equation (12) is at first solved on the assumption that $M=0$; in this case

$$u_0=C_1g(r)+C_2h(r)+\gamma mr^2/2kT$$

where C_1 and C_2 are constants and the functions $g(r)$ and $h(r)$ are solutions of the equation

$$r^{-2}(d/dr)r^2\,du/dr-(d\Phi_0/kTdr)(du/dr)-6u/r^2=0 \quad . \quad . \quad . \quad . \quad (15)$$

$g=0$ for $r=0$; $h=0$ for $r\to\infty$. It was found that a constant a can be defined so that

$$\lim_{r\to\infty} g''=a\lim_{r=0} g''. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (16)$$

[In eqns. (16), (18), (22), (23) differentiation is indicated by a dash.]

In those terms of eqn. (12) which are proportional to M^2, a linear combination of the functions $u_0(r)$ and $g(r)$ is now substituted for the unknown $u_M(r)$ so that the solution of (12) which satisfies condition (14) is completely specified in terms of known functions

$$u_M=u_0+(M^2/kT)\left\{C_5g+C_6h+\int_B^r G(s)[g(r)h(s)-h(r)g(s)]s^2\,ds\right\} \quad (17)$$

where
$$G(r)=(\gamma m/3kT)[\Phi_1'+(2\Phi_2'/7)-(18\Phi_2/7r)] +(u_0'+C_3gg')(\Phi_1'+2\Phi_2'/7)+(u_0+C_3g)(6\Phi_2/7r^2) \quad . \quad (18)$$
and
$$C_3=4\gamma ma/kT \quad C_5=\int_B^\infty G(s)f_0(s)h(s)s^2\,ds \quad C_6=-\int_B^\infty G(s)f_0(s)g(s)s^2\,ds. \quad . \; . \; . \quad (19)$$

§ 5. THE INCREMENT IN VISCOSITY

The coefficient of viscosity, η, is equal to
$$\eta=-(N^2/2AV)\int(\partial\chi_M/\partial z)yf_MW_M\,d\mathbf{r} \quad . \; . \; . \; . \quad (20)$$
and $\varDelta\eta$, the increment which is due to the electric field is given by
$$\varDelta\eta/\eta=Q_M/Q \quad . \; . \; . \; . \; . \; . \; . \; . \; . \; . \quad (21)$$
where
$$Q_M=\int_B^\infty[(u_M-u_0)\chi_0'+(12M^2u_0/7r^4)-(2M^2\Phi_2'\chi_0'u_0/7k^2T^2)]f_0r^3\,dr \quad . \; . \; . \quad (22)$$
$$Q=\int_B^\infty \chi_0'u_0f_0r^3\,dr. \quad . \; . \; . \; . \; . \; . \; . \; . \quad (23)$$

Expression (21) was evaluated for chloroform. It was assumed that the radial distribution f_0 is similar to that of monatomic liquids having a maximum (at $r=R_1$), a minimum (at $r=R_2>R_1$) and then approaches gradually a constant value. A distribution function was plotted so that the value of R_1 was in accordance with the mean distance between molecules as derived from the density and that the maximum of f_0 corresponded to 10 nearest neighbours for a molecule. The function Φ_0 which is derived from this distribution is specified in the first row of the table. The justification for the use of an almost arbitrary distribution is given in the next section.

No account was taken of the possible effect of the field on the friction constant γ. Since this constant is closely related to the mean square force in thermal equilibrium and the latter is in first approximation independent of the field strength it is not likely that, in neglecting the field effect on γ, an appreciable error was introduced.

§ 6. NUMERICAL TESTS

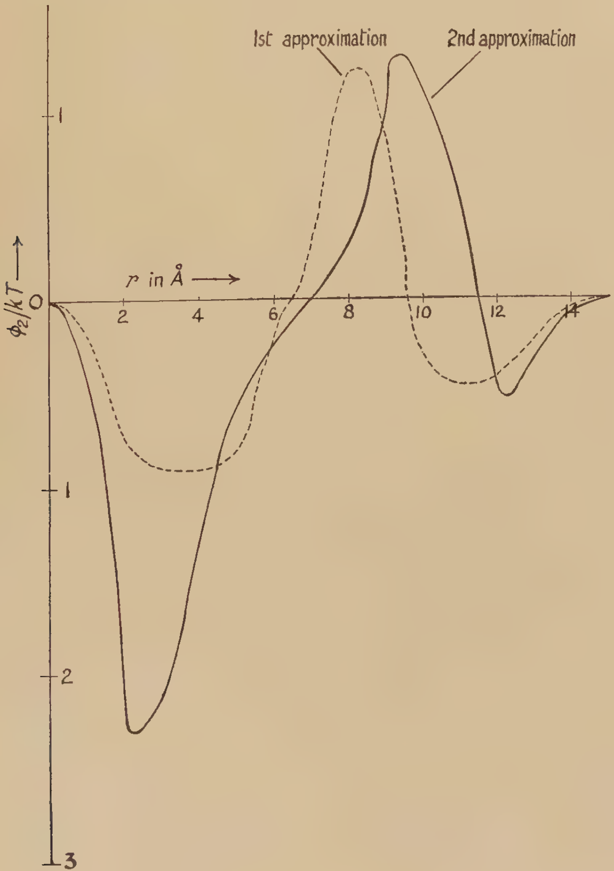
The calculated value of $\varDelta\eta/\eta F^2$ is virtually independent of the exact shape of the Φ_0-curve. This was shown by calculations made with a number of different Φ_0-curves characteristics of which are given in the table. The viscosities calculated in these tests varied by 1/600 whereas the values of $\varDelta\eta/\eta F^2$ were changed by hardly more than 20%.

The two approximations to Φ_0 (eqns. (7) and (10)) were compared by calculating the function Φ_2, using the data of the first row of the table. Result (fig. 1) shows that the difference is not excessive. The values of $\varDelta\eta/\eta F^2$ as obtained with the different approximations have a ratio of 1·33.

The calculated relative increment in viscosity is accordingly not very sensitive towards the details of the calculation and essentially determined by the induced forces of molecular interaction.

R_1 Å	R_2 Å	$\frac{-\Phi_0(R_1)}{kT}$	$\frac{\Phi_0(R_2)}{kT}$	$r>R_2$ $\frac{10^{-4}\,r^7}{kT}$ r in Å	$\frac{2V10^{-10}\eta kT}{m\gamma N^2}$	$\frac{10^8\Delta\eta}{\eta F^2}$
5·5	8·25	3·0	1·0	430	687	2·4
3·75	4·75	1·48	0·85	graph	13·4	2·81
3·75	4·75	1·48	0·85	$1{\cdot}2r$	59·8	3·1
3·75	4·75	1·48	0·85	5·4	16·5	2·9
3·75	4·75	1·48	0·85	$29r^{-3}$	5·15	2·6
3·75	4·75	1·2	0·64	4·76	4·71	2·7
3·75	4·75	1·2	0·74	5·1	9·12	2·8
3·75	4·75	1·2	0·38	2·96	1·56	2·35
3·95	4·95	0·93	0·54	7·9	1·56	2·5
3·75	4·75	0·93	0·54	3·6	3·35	2·5

Fig. 1



§7. COMPARISON WITH EXPERIMENT

Using the Φ_0-curve as specified in the first row of the table, the values $M/F=1{\cdot}10\times10^{-22}$ e.s.u., $\epsilon=5{\cdot}05$ and eqn. (10) for Φ_M it is found that

$$\Delta\eta/\eta F_e^2=20{\cdot}8\times10^{-8}\text{ e.s.u.} \qquad (24)$$

comparing with the experimental 'viscoelectric constant' of $17{\cdot}0\times10^{-8}$ e.s.u. for chloroform. Sign and order of magnitude are accordingly in agreement with experiment.

Further experimental results are available for chlorobenzene and amyl acetate and do not differ very much from the figure given for chloroform. It would appear that the theory is well compatible with these measurements although no calculations were made.

The success of the theory is qualified by the absence of any observable effect of the field on the viscosity of benzene, the only non-polar liquid investigated by Andrade and Dodd. The corresponding viscoelectric constant cannot be larger than 10^{-9} whereas the theory should yield a value much smaller than for chloroform but still about 10^{-8}.

This experimental result suggests an explanation of the field effect on viscosity in which the difference of non-polar and polar liquids is emphasized and the non-spherical shape of polar molecules plays an important part. There can, however, be no gross miscalculation in the present estimate of the induced forces and, whatever alternative influences may be important, it would be difficult to understand why the induced forces fail to show in viscosity.

Discrepancies would be resolved if there are forces, so far not taken into account, which counteract the forces that have been considered. The repulsive overlap forces may be of this kind, since the electronic wave functions are extended in direction of the field and, correspondingly, contracted in perpendicular directions. The exchange integral accounting for repulsion should, accordingly, be increased near the induced poles and reduced about the equator thus accounting for the behaviour of benzene. (The total induced energy and, accordingly, the internal field strength are not appreciably affected on account of the short range of the repulsive forces.) A corresponding reduction of the viscoelectric constant of chloroform would be partly offset by the effect of its non-spherical shape so that the above eqn. (24) may be regarded as valid.

ACKNOWLEDGMENTS

We wish to express our thanks to the University of London Central Research Fund for supplying a 'Facit' calculator which was used for numerical work in connection with this paper. One of us (G. H. A. C.) gratefully acknowledges the maintenance grant from the Department of Scientific and Industrial Research which he received during the course of this work.

REFERENCES

ANDRADE, E. N. da C., and DODD, C., 1950, *Proc. Roy. Soc.* A, **204**, 449.
DEBYE, P., 1912, *Phys. Z.*, **13**, 97.
EISENSCHITZ, R., 1949, *Proc. Phys. Soc.* A, **62**, 41.
KIRKWOOD, J. G., 1946, *J. Chem. Phys.*, **14**, 180.

XLVIII. *On the Molecular Distribution Functions of a One-Dimensional Fluid: I*

By Alladi Ramakrishnan
Department of Physics, University of Madras*

[Received January 12, 1954]

Summary

The results of Salzburg, Zwanzig and Kirkwood relating to the molecular distribution functions of a one-dimensional fluid are obtained as corollaries of a recent contribution by the author and P. M. Mathews (1953) to this Magazine. These functions are identified as product densities and the problem is considered from the standpoint of stochastic theory.

§ 1. Introduction

Recently attempts have been made by many authors (see Born and Green 1949) to develop a kinetic theory of liquids based on molecular distribution functions (they were first referred to as multiform distribution functions by Born and Green to distinguish them from the familiar probability distribution functions). In order to understand the nature of the mathematical problems presented by this new theory, Salzburg, Zwanzig and Kirkwood (SZK, 1953) studied the case of a one-dimensional fluid. In this paper we shall show that a proper interpretation of these functions from the standpoint of the theory of stochastic processes not only rationalizes the method of SZK but helps us to obtain their results in the case of a one-dimensional fluid in a very easy manner by a new mathematical approach which, it is hoped, will yield fruitful results even in more complicated cases.

In a series of papers† (A, B, C, D) the author formulated and developed " the theory of product densities " in dealing with a stochastic variable representing the number of ' particles ' or ' entities ' distributed in a continuous infinity of states. In Paper D a class of product densities was considered and we shall now show that *the problem of the one-dimensional fluid if suitably interpreted can be solved as a corollary of the results obtained in that paper.* For this purpose we shall formulate and solve the following problem of random points in a line.

---

* Communicated by the Author.

† Alladi Ramakrishnan, 1950, *Proc. Camb. Phil. Soc.*, **46,** 595 ; 1952, *Ibid.*, **48,** 451 ; 1953, *Ibid.*, **49,** 473 ; Alladi Ramakrishnan and P. M. Mathews, 1953, *Phil. Mag.*, **44,** 1122.

These papers will hereafter be referred to as Papers A, B, C and D respectively. The notation here corresponds to that used in Paper D, but no attempt is made to keep it in conformity with those of A, B and C though we make use of the results derived therein.

§ 2. RANDOM POINTS IN A LINE

Consider points randomly distributed on a line represented by the parameter t in the following manner.

The conditional probability that there is no random point between $t=t_1$ and $t=t_2$ $(t_2>t_1)$, given that there is a point at $t=t_1$, is $\phi(t_2-t_1)$ and is independent of the distribution of points at parametric values $t<t_1$. It was shown in Paper D that this function uniquely determines the statistical distribution of the points on the line. Let $\pi(n, t)$ be the probability that there are n random points between $t=0$ and t given that there is a point at $t=0$. Using the ' method of regeneration points ' the author obtained the integral equation

$$\pi(n, t)=-\int_0^t \pi(n-1, t-\tau)\phi'(\tau)\, d\tau+\delta(n)\phi(t), \quad . \quad . \quad . \quad (1)$$

where $\phi'(\tau)=d\phi(\tau)/d\tau$ and $\delta(n)=1$ for $n=0$ and is zero otherwise. We shall now deduce some corollaries of the above equation :

If $\omega(n, t)$ is the probability that the nth point (omitting the point at $t=0$) lies between t and $t+dt$ given that at $t=0$ there is a point, then $\omega(n, t)$ satisfies the integral equation

$$\omega(n, t)=\int_0^t \omega(n-1, t-\tau)\omega(\tau)\, d\tau+\delta(n-1)\omega(t), \quad . \quad . \quad . \quad (2)$$

where $\omega(\tau)\equiv \omega(1, \tau)=-\phi'(\tau)$.

If $\sigma(s)$ and $p(n, s)$ are the Laplace Transforms* of $\phi(t)$ and $\pi(n, t)$ then it was proved in paper D that

$$p(n, s)=\sigma(s)[1-s\sigma(s)]^n. \quad . \quad . \quad . \quad . \quad . \quad . \quad (3)$$

If we define $\eta(n, s)$ as the L.T. of $\omega(n, t)$ and write $\eta(s)$ for $\eta(1, s)$, we obtain from (2)

$$\eta(n, s)=[1-s\sigma(s)]^n=[\eta(s)]^n. \quad . \quad . \quad . \quad . \quad (4)$$

From (4) it follows that

$$\omega(n, t)=\int_0^t\int_{t_1}^t \dots \int_{t_{n-2}}^t \omega(t_1)\omega(t_2-t_1)\dots \omega(t-t_{n-1})\, dt_1\, dt_2 \dots dt_{n-1} \quad (5)$$

i.e. $\omega(n, t)$ is the n-fold convolution of the function $\omega(t)$ where $\omega(t)\, dt$ is the probability that the ' first ' point (counted from $t=0$) occurs between t and $t+dt$ given that a point (uncounted) occurs at $t=0$.

We shall call $\omega(t)$ the ' *basic function* ' of the random distribution of points. We shall show that it is $\omega(n, t)$ and not $\pi(n, t)$ that plays a fundamental role in the kinetic theory of a one-dimensional fluid. We note that $\omega(n, t)$ unlike $\pi(n, t)$ is a probability density with respect to t and $\int_0^\infty \omega(n, t)dt=1$ for all n.

---

* In this paper, L.T. means Laplace Transform.

We obtain from (3) and (4)

$$sp(n, s)=\eta(n, s)-\eta(n+1, s)$$

i.e.
$$\frac{\partial\pi(n, t)}{\partial t}=\omega(n, t)-\omega(n+1, t). \qquad (6)$$

This equation can also be obtained by elementary probability arguments.

If $f_n^{(F)}(t_1, t_2, \ldots, t_n)$* is the product density of degree n of random points, given that there is a point at $t=0$, the nature of the random distribution we have assumed implies that

$$f_n^{(F)}(t_1, t_2, \ldots, t_n)=f_1^{(F)}(t_1)f_1^{(F)}(t_2-t_1)\ldots f_1^{(F)}(t_n-t_{n-1}), \qquad (7)$$

provided $t_1, t_2, \ldots, t_n$ are ordered. As stated in Paper D if $t_1, t_2, \ldots, t_n$ are not ordered, $f_n^{(F)}$ still exists. $f_n^{(F)}$ by definition is symmetric in the variables $t_1, t_2, \ldots, t_n$. We can therefore order them and then apply (7).

By simple reasoning we note that

$$f_1^{(F)}(t)=\sum_{n=1}^{\infty}\omega(n, t). \qquad (8)$$

Therefore if $\psi(s)$ is the L.T. of $f_1^{(F)}(t)$

$$\psi(s)=\sum_{n=1}^{\infty}[\eta(s)]^n=\frac{\eta(s)}{1-\eta(s)}=\frac{1-s\sigma(s)}{s\sigma(s)}. \qquad (9)$$

For reasons which will be obvious presently we shall call the above distribution of points on the line *a 'free' distribution with a basic function* $\omega(t)$. Stochastic processes associated with such a distribution in the special case when $\omega(t)=e^{-\lambda t}\lambda$ i.e. where $\pi(n, t)$ is Poissonian were discussed in Paper C.

Now let us set ourselves the following problem : Given that the $N+1$th particle is at $t=L$, what is the nature of the distribution of the N particles between 0 and L ?

For convenience we shall call this conditional distribution *'the constrained' distribution with the basic function* $\omega(t)$. For our purpose it is sufficient if we are able to determine $f_n^{(c)}$, the product density of degree n of the 'constrained' distribution. By direct probability arguments we obtain

$$f_n^{(c)}(t_1, t_2, \ldots, t_n ; L)$$
$$=\sum_{m_i}\frac{\omega(m_1, t_1)\omega(m_2, t_2-t_1)\ldots\omega(m_n, t_n-t_{n-1})\omega(m_{n+1}, L-t_n)}{\omega(N+1, L)}, \qquad (10)$$

where $\sum\limits_{m_i}$ indicates summation with respect to all m_i's $(i=1, 2, \ldots, n+1)$ over all positive integral values of $m_i (m_i\neq 0)$ such that $\sum\limits_{i=1}^{n+1} m_i=N+1$.

We shall show in § 3 that this is equivalent to the expression for $f_n^{(c)}$ obtained by SZK by a tedious process of integration.

---

* In this paper f_n denotes a product density of degree n. The superscripts are used to distinguish between various product-densities of the same degree. For the definition and properties of product densities see Papers A and B.

We note two important features characteristic of $f_n^{(c)}$.

(1) $f_n^{(c)}$ is no longer related in a simple manner to $f_1^{(c)}$ as $f_n^{(F)}$ is related to $f_1^{(F)}$.

(2) Though $f_n^{(c)}$ is uniquely 'generated' by $\omega(m, t)$ the converse is not true. In (10), let us replace $\omega(m, t)$ by another function $Q(m, t)$ which 'generates' the same $f_n^{(c)}$. We can always write

$$Q(m, t)=q(m, t)\omega(m, t). \tag{11}$$

Then we find that

$$q(m_1, t_1)q(m_2, t_2-t_1)\dots q(m_n, t_n-t_{n-1})q(m_{n+1}, L-t_n)=q(N+1, L). \tag{12}$$

Since $\sum_{i=1}^{n+1} m_i=N+1$ and L is the sum of the intervals $t_1, t_2-t_1, \dots, L-t_n$, it follows that

$$q(m, t)=\exp(\mu m+\lambda t), \tag{13}$$

where μ and λ are arbitrary constants. We can express (11) in the following form:

Lemma I:

If $Q(m, t)$ *generates* $f_n^{(c)}$, *then* $\exp(\mu m+\lambda t)Q(m, t)$ *also generates* $f_n^{(c)}$.

Since $\omega(n, t)$ is a probability density, if we set

$$\omega(n, t)=\exp[-(\mu n+\lambda t)]Q(n, t), \tag{14}$$

$$\int_0^\infty \exp[-(\mu n+\lambda t)]Q(n, t)\,dt=1, \quad \text{or} \quad \exp(\mu n)=\int_0^\infty \exp(-\lambda t)Q(n, t)\,dt. \tag{15}$$

If $\Omega(n, s)$ is the L.T. of $Q(n, t)$ from (14) we obtain

$$\Omega(n, s)=\exp(\mu n)\eta(n, s-\lambda)=[\exp(\mu)\eta(s-\lambda)]^n=[\Omega(1, s)]^n=[\Omega(s)]^n. \tag{16}$$

Hence writing

$$Q(t)\equiv Q(1, t), \tag{17}$$

if follows that

$$Q(n, t)=\int_0^t\int_{t_1}^t\dots\int_{t_{n-2}}^t Q(t_1)Q(t_2-t_1)\dots Q(t-t_{n-1})\,dt_1\,dt_2\dots dt_{n-1}. \tag{18}$$

Since $\omega(t)$ is a probability density

$$\int_0^\infty \omega(t)\,dt=\int_0^\infty \exp[-(\mu+\lambda t)]Q(t)\,dt=1, \quad \text{i.e.} \exp(\mu)=\Omega(\lambda), \tag{19}$$

and

$$\eta(s)=\frac{\Omega(s+\lambda)}{\Omega(\lambda)}, \quad \Omega(s)=\Omega(\lambda)\eta(s-\lambda). \tag{20}$$

Lemma II:

The basic function $\omega(t)$ *can always be obtained from any* $Q(t)$ *where* $Q(n, t)$, *the n-fold convolute of* $Q(t)$ *generates the 'constrained' distribution.*

We note that the basic function contains one arbitrary constant λ which we shall call the parameter of the 'free' distribution.

Our fundamental equation (10) for $f_n^{(c)}$ can be written down directly by simple probability arguments only when we deal with the basic function. SZK, on the contrary, dealt with a $Q(t)$ which had no probability significance and so they were compelled to obtain $f_n^{(c)}$ from $f_N^{(c)}$ by a process of integration.

A simple corollary of our eqn. (10) is the following. If $\omega^{(c)}(n, t, L)\,dt$ is the probability that the nth particle occurs between t and $t+dt$ given that the $N+1$th lies at L then we obtain

$$\omega^{(c)}(n, t, L)=\frac{\omega(n, t)\omega(N+1-n, L-t)}{\omega(N+1, L)}, \quad . \quad . \quad . \quad (21)$$

and

$$\frac{\partial\pi^{(c)}(n, t, L)}{\partial t}=\omega^{(c)}(n, t, L)-\omega^{(c)}(n+1, t, L), \quad . \quad . \quad . \quad (22)$$

where $\pi^{(c)}(n, t, L)$ is the probability that n particles lie between 0 and t given that the $N+1$th particle is at L. There is of course a particle at $t=0$ and this is not counted.

§ 3. Application to the Theory of a One-Dimensional Fluid

SZK assumed an interaction only between the nearest neighbours of a one-dimensional fluid. They defined $P_N(t_1, t_2, \ldots, t_N)\,dt_1\,dt_2\ldots dt_N$ as the probability that there is a particle between t_1 and t_1+dt_1 one between t_2 and $t_2+dt_2, \ldots,$ and one between t_N and t_N+dt_N given that there is a particle (uncounted) at $t=0$ and the $N+1$th particle is at L (this takes care of wall effects). They wrote

$$P_N(t_1, t_2, \ldots, t_N)=\exp(\beta\bar{A}-\beta[V(t_1)+V(t_2-t_1) + \ldots +V(t_N-t_{N-1})+V(L-t_N)]), \quad . \quad . \quad . \quad (23)$$

where

$$\exp(-\beta\bar{A})=\int_0^L\int_0^L\ldots\int_0^L \exp(-\beta[V(t_1)+V(t_2-t_1) +\ldots+V(t_N-t_{N-1})+V(L-t_N)])\,dt_1\,dt_2\ldots dt_N. \quad . \quad . \quad . \quad (24)$$

Though it may seem trivial, the author wishes to stress that P_N is a product density and hence if there are N particles between 0 and L the integral with respect to each of the variables $t_1, t_2, \ldots, t_N$ over the whole domain 0 to L should yield $N\,!$ and not 1 as it does according to (23). P_N should be identical with $f_N^{(c)}$ which according to (10) should be given by

$$P_N\equiv f_N^{(c)}(t_1, t_2, \ldots, t_N) =\frac{\exp(-\beta[V(t_1)+V(t_2-t_1)+\ldots+V(t_N-t_{N-1})+V(L-t_N)])}{Q(N+1, L)}, \quad (25)$$

where

$$Q(N+1, L)=\int_0^L\int_{t_1}^L\ldots\int_{t_{N-1}}^L \exp(-\beta[V(t_1)+V(t_2-t_1) +\ldots+V(t_N-t_{N-1})+V(L-t_N)])\,dt_1\,dt_2\ldots dt_N. \quad . \quad . \quad (26)$$

In spite of this, SZK obtained correct expressions for $f_n^{(c)}$ (which they denoted by $\rho^{(n)}$) because they consistently integrated expressions involving the numerator of their P_N over the whole domain thus introducing a factor $N\,!$ in the numerator, which annulled the defect in their definition of P_N. We will rationalize their process in § 4. Comparing the corrected expression for $P_N \equiv f_N^{(c)}$ with (10) we find

$$Q(t) \equiv \exp[-\beta V(t)]. \qquad (27)$$

Thus the basic function is given by

$$\omega(t) = \exp\left(-[\mu + \lambda t + \beta V(t)]\right) \qquad (28)$$

where

$$\exp(\mu) = \int_0^\infty \exp\left(-[\lambda t + \beta V(t)]\right) dt. \qquad (29)$$

We note that μ is a function of λ and hence our expression for the basic function ω contains one arbitrary constant which is consistent with the statement that the potential V contains an arbitrary constant. Thus the expression for $f_n^{(c)}$ given by SZK is identical with (10). The corrected expression is $P_N \equiv f_N^{(c)} \equiv \rho^{(N)}$.*

Now let us consider the following potential V:

$$\begin{aligned} V(t) &= \infty, \qquad 0 < t < a \\ V(t) &= 0, \qquad t > a \end{aligned} \qquad (30)$$

i.e.

$$\exp[-V(t)] = H(t-a), \qquad (31)$$

where $H(x)$ is the Heaviside unit function, i.e. $H(x)=1$ if $x>0$, $H(x)=0$ if $x<0$.

$$\omega(t) = H(t-a)\lambda \exp[-\lambda(t-a)], \qquad -\mu = \log\lambda + a\lambda \qquad (32)$$

$$\omega(n, t) = H(t-na) \exp[-\lambda(t-na)] \frac{(t-na)^{n-1}}{(n-1)\,!} \lambda^n. \qquad (33)$$

$f_n^{(c)}$ is given by applying (10). In particular

$$\rho^{(1)}(t) = f_1^{(c)}(t) = \sum_{n=1}^{N} \frac{N\,!}{(n-1)\,!\,(N-n)\,!} \frac{(t-na)^{n-1}\,(L-t-\overline{N+1-na})^{N-n}}{(L-\overline{N+1}\,a)^N}$$
$$\times H(t-na) H(L-t-\overline{N+1-na}). \qquad (34)$$

§ 4. INTEGRATION PROCESS OF SZK

We shall now discuss the integration process of SZK. For this purpose we have to recall another species of distribution function—the Janossy density defined in Paper B. Adapting the definition of Janossy density to our problem we say $J_n(t_1, t_2, \ldots, t_n)\, dt_1\, dt_2 \ldots dt_n$ is the probability that there is one particle in dt_1, one in $dt_2, \ldots$, and one in dt_n and *none* elsewhere. We quote here a result proved in paper B connecting product densities and the Janossy densities:

$$f_n(t_1, t_2, \ldots, t_n) = \sum_{m=n}^{\infty} \frac{1}{(m-n)\,!} \int_0^L \int_0^L \cdots \int_0^L J_m(t_1, t_2, \ldots, t_m)$$
$$\times dt_{n+1}\, dt_{n+2} \ldots dt_m. \qquad (35)$$

---

* If we use the SZK definition of P_N, $\rho^{(N)}$ will not be identical with P_N and this is illogical.

If we are given that the $N+1$th particle occurs at L the Janossy densities of all orders except $m=N$ vanish and

$$\rho^{(n)}=f_n^{(c)}(t_1, t_2, \ldots, t_n)=\frac{1}{(N-n)\,!}\int_0^L\int_0^L\cdots\int_0^L J_N^{(c)}(t_1, t_2, \ldots, t_N)$$
$$\times\, dt_{n+1}\,dt_{n+2}\ldots dt_N. \qquad \ldots\ldots\ldots \qquad (36)$$

We also note that $J_N^{(c)}\equiv f_N^{(c)}$ and this is identical with our corrected definition for P_N. The most interesting feature of $J_N^{(c)}$ is that it is symmetric in the N variables $t_1, t_2, \ldots, t_N$ and exists even when they are unordered but can be explicitly written down according to (25) *only after we order them.* Hence the computation of the multiple integral is complicated and is done in the following manner.

We first order the $N-n$ variables $t_{n+1}, t_{n+2}, \ldots, t_N$ not concerning ourselves with their order relative to the other n variables $t_1, t_2, \ldots, t_n$. We rewrite eqn. (36) using the symmetry property of $f_n^{(c)}$ as

$$f_n^{(c)}(t_1, t_2, \ldots, t_n)$$
$$=\int_0^L\int_{t_{n+1}}^L\cdots\int_{t_{N-1}}^L J_N^{(c)}(t_1, t_2, \ldots, t_N)\,dt_{n+1}\,dt_{n+2}\ldots dt_N. \qquad \ldots \qquad (37)$$

Let us now assume $t_1, t_2, \ldots, t_n$ are ordered. $J_N^{(c)}$ cannot yet be explicitly written down unless the order of $t_1, t_2, \ldots, t_n$ relative to the other $N-n$ ordered variables $t_{n+1}, t_{n+2}, \ldots, t_N$ is known. The relative order changes in the process of integration and so to evaluate the integral (37), SZK adopted the ingenious device of writing (37) in the form

$$f_n^{(c)}(t_1, t_2, \ldots, t_n)=\int_0^L\int_{\tau_1}^L\int_{\tau_2}^L\cdots\int_{\tau_{N-1}}^L \sum_k J_N(\tau_1, \tau_2, \ldots, \tau_N).$$
$$\delta(\tau_{k_1}-t_1)\delta(\tau_{k_2}-t_2)\ldots\delta(\tau_{k_n}-t_n)\,d\tau_1\,d\tau_2\ldots d\tau_N. \qquad \ldots \qquad (38)$$

$\sum\limits_k$ denotes summation over all values of $k_1, k_2, \ldots, k_n$ subject to the condition $k_1<k_2<\ldots<k_n$. Since according to (25), $J_N^{(c)}$ is given in terms of $Q(\tau)=\exp\left[-\beta V(\tau)\right]$, this process yields our fundamental eqn. (10) but with $Q(n, t)$ in the place of $\omega(n, t)$. Though SZK used the uncorrected expression for $J_N^{(c)}\equiv P_N$ in (44) they were led to the correct result since in (44) they integrated with respect to each of the N variables *over the whole domain* 0 to L thus introducing the factor $N\,!$ which annulled the defect in their definition of P_N.

§5. ASYMPTOTIC FORMS OF $f_n^{(c)}\equiv\rho^{(n)}$

If $N\gg 1$; $L\gg t, a$ (34) takes the asymptotic form*

$$\rho^{(1)}(t)\sim\sum_{n=1}^{\infty}H(t-na)\frac{\exp\left[-\dfrac{N(t-na)}{L-Na}\right]}{(n-1)\,!}\left[\frac{N\,(t-na)}{L-Na}\right]^{n-1}\frac{N}{L-Na}. \qquad . \qquad (39)$$

---

* We shall not bother ourselves with a formal derivation of the asymptotic form which can be obtained in just the same manner as a Poisson approximation from a binomial distribution.

We can also write the asymptotic expression for $\rho^{(n)}$ as

$$\rho^{(n)}(t_1, t_2, \ldots, t_n) \sim \rho^{(1)}(t_1)\rho^{(1)}(t_2-t_1) \ldots \rho^{(1)}(t_n-t_{n-1}), \quad . \quad . \quad (40)$$

provided $N \gg 1$; $L \gg t_1, t_2-t_1, t_3-t_2, \ldots, t_n-t_{n-1}$ and $L-t_n \gg a$.

Comparing (39) and (40) with the corresponding expressions in the case of a ' free ' distribution we are led to the following result :

Lemma III :

In the case of a potential of width a and infinite height if $N \gg 1$ and $L \gg a$, as long as we are dealing with the distribution of particles in a domain 0 to t where $L \gg t$, we can replace the ' constrained ' distribution by a ' free ' distribution with the parameter $\lambda = \rho = N/(L-Na)$.

We therefore write, using (22) and (33)

$$\pi^{(c)}(n, t, L) = \int_0^{t-na} \lambda^n \exp(-\lambda t) \frac{t^{n-1}}{(n-1)\,!}\, dt$$

$$- \int_0^{t-(n+1)a} \lambda^{n+1} \exp(-\lambda t) \frac{t^n}{n\,!}\, dt, \quad \lambda = \frac{N}{L-Na} . \quad . \quad . \quad (41)$$

SZK obtained the asymptotic expression for $\rho^{(2)}(t_1, t_2)$ in the case when $t_1 \gg a$, but t_2-t_1 is comparable to a. They wrote $\rho^{(1)}(t_1) \sim N/L$ for $t_1, L-t_1 \gg a, t_2-t_1$. The author is not in agreement with SZK in regard to this value of $\rho^{(1)}(t_1)$ and wishes to point out that $\rho^{(1)}(t_1) \sim N/L-Na$ for t_1, $L-t_1 \gg a$. A comparison between the asymptotic forms of SZK and that of the author will be made in a later paper in which he also proposes to discuss the molecular distribution in the case of a square well potential defined by

$$\begin{aligned} V(t) &= \infty, && 0<t<a \\ V(t) &= -\epsilon, && a<t<b \\ V(t) &= 0, && t>b \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (42) \end{aligned}$$

by using the method of basic functions. In that paper it will be shown that if the basic function $\omega(t)$ is discontinuous at a finite number of points, it leads to difference differential equations which can be completely solved. The problem of a one-dimensional gas with potential functions defined by (42) was proposed and discussed recently be Sells, Harris and Guth (1953).

§ 6. SOME REMARKS ON PRODUCT DENSITIES

Since molecular distribution functions have been identified as product densities, it is relevant to note the following properties of product density functions in addition to those stated and proved in Papers A to D.

(1) Product densities of any degree are defined with respect to a continuum, which in general can be n-dimensional. If a product density is integrated with respect to r dimensions it is still a product-density with respect to the continuum of $n-r$ dimensions.

(2) Taking in general a product density of degree h as $f_h(t_1, t_2, \ldots, t_h)$[*] let us ask the question : What is the effect of integrating f_h with respect to only one variable ? Writing

$$\int_{t_h} f_h(t_1, t_2, \ldots, t_h)\, dt_1\, dt_2 \ldots dt_h = R_{h-1}(t_1, t_2, \ldots, t_{h-1})\, dt_1\, dt_2 \ldots dt_{h-1}, \qquad \ldots \quad (43)$$

this represents the expectation value of the product of the number of particles in the range of integration of t_h and the number of particles in $dt_1, dt_2, \ldots, dt_{h-1}$. Then R_{h-1} is no longer a product–density and $R_{h-1}\, dt_1\, dt_2 \ldots dt_{h-1}$ no longer a probability magnitude.

In particular when we have a total number N and only N particles in the entire range,

$$R^{(N)}_{h-1}(t_1, t_2, \ldots, t_{h-1}) = (N-h+1) f^{(N)}_{h-1}(t_1, t_2, \ldots, t_{h-1}), \qquad (44)$$

if f_h is integrated over the whole range of t_h since the number of particles outside the infinitesimal ranges $dt_1, dt_2, \ldots, dt_{h-1}$ is $N-h+1$ *conditional* upon the existence of particles in $dt_1, dt_2, \ldots, dt_{h-1}$ and so it is not a random variable. From (44) we immediately get

$$f^{(N)}_h(t_1, t_2, \ldots, t_h) = \frac{1}{(N-h)\,!} \int_{t_{h+1}} \int_{t_{h+2}} \ldots \int_{t_N} f^{(N)}_N(t_1, t_2, \ldots, t_N) \times dt_{h+1}\, dt_{h+2} \ldots dt_N. \qquad \ldots \ldots \quad (45)$$

Born and Green state that (44) is obvious but it is not clear why it is so unless we interpret their functions as product densities. Since such functions were not so identified and distinguished from probability densities, in the case of the ‘ distribution functions of many electron systems ’, Mott and Sneddon (1950) had to appeal to an artificial rule of integration. They stated that to obtain $f^{(N)}_h$ from $f^{(N)}_N$ we should integrate $f^{(N)}_N$ with respect to $t_{h+1}, t_{h+2}, \ldots, t_N$ and the “ integration must be carried out in such a way that each configuration must appear only once ; that is, $dt_{h+1}\, dt_{h+2} \ldots dt_N$ must be replaced by

$$\frac{dt_{h+1}\, dt_{h+2} \ldots dt_N}{(N-h)\,!} \text{”}.$$

This argument is not correct and must be replaced by the mathematical arguments outlined above.

REFERENCES

BORN, M., and GREEN, H. S., 1949, *A General Kinetic Theory of Liquids* (Cambridge : University Press).

MOTT, N. F., and SNEDDON, I., 1950, *Wave Mechanics and its Applications* (Oxford).

SALZBURG, Z. W., ZWANZIG, R. W., and KIRKWOOD, J. G., 1953, *J. Chem. Phys.*, **21**, 1098.

SELLS, R. L., HARRIS, C. W., and GUTH, E., 1953, *J. Chem. Phys.*, **21**, 1617.

---

[*] Each of the variables $t_1, t_2, \ldots, t_h$ has n components if the t-space is n-dimensional. Integration with respect to a variable, i.e. with respect to each of the n components of that variable, should be distinguished from integration over a dimension which means integration with respect to the corresponding components of all the variables.

XLIX. *Isolation and Measurement of the Helium-*4, *Helium-*3 *and Tritium Produced in Steel by* 340 MEV *Proton Bombardment*

By G. R. MARTIN, S. J. THOMSON and G. WARDLE
Londonderry Laboratory for Radiochemistry, University of Durham
and
K. I. MAYNE
Clarendon Laboratory, Oxford*

[Received January 8, 1954]

SUMMARY

The small amounts of ^{4}He, ^{3}He, and ^{3}H, produced in a block of steel bombarded with 340 MEV protons have been isolated. Measurements have been made of the total helium by a gas-volumetric method, of the tritium by counting of the associated β-particles, and of the ^{3}He : ^{4}He ratio by mass-spectrometry. The ratio ^{4}He : ^{3}He : ^{3}H has been found to be 1·00 : 0·048 : 0·078 ; this result is discussed in the light of Le Couteur's theory of nuclear evaporation.

INVESTIGATIONS of the nuclear evaporation processes induced by bombardment with particles of very high energy have, so far, mostly been made by one or other of two complementary techniques : by radiochemical studies of the yields of radioactive spallation products remaining after the completion of the evaporation process, and by the examination of the tracks produced in photographic plates by the charged particles which have been emitted.

The correlation of the resulting data is rendered difficult by two facts. In the first place, ‘ evaporation stars ’ in photographic plates must, in general, have originated in one of the elements (H, C, N, O, Br, Ag) naturally present in the emulsions. This immediately restricts the range of possible comparisons and, further, makes it difficult to obtain an unbiased selection of ‘ stars ’ without introducing some uncertainty concerning the origin of the events. It therefore seemed desirable to make a comparison of the light particles evaporated with the spallation products from the same material under similar conditions of bombardment. Secondly, although it is comparatively easy to distinguish between singly and doubly charged particles, mass discrimination is by no means so readily achieved. Most of the attempts in this direction have involved

---

* Communicated by the Authors.

some inherent bias, introduced, for example, by the necessity to select tracks of length sufficient for the measurement of the mean deviation from a rectilinear path (see, for example, Holtebekk, Isachsen and Sörensen 1953).

In the present work we have made a direct measurement of the total yields of three of the evaporation particles (^{4}He, ^{3}He and ^{3}H) produced in a sample of steel which had been bombarded with 13 μA-hr. of 340 MeV protons in the Berkeley synchro-cyclotron.

Total helium has been measured gas-volumetrically in an apparatus similar to that recently described by Glückauf (Glückauf 1946, Chackett, Reasbeck and Wilson 1953), tritium has been estimated by gas counting of the β-particles emitted (Allen and Ruben 1942, Libby and Barter 1942), and the helium isotope ratio determined by mass-spectrometry in a high resolution Nier-type instrument (Mayne 1954). Helium was extracted by solution of the steel in aqueous copper potassium chloride solution; this resulted in an excessive dilution of the tritium with ordinary hydrogen, and the release of the tritium was therefore achieved in the dry way. Vacuum fusion, and ignition of the steel in an atmosphere of chlorine were both used, and gave substantially the same result. Since it was thus not feasible to measure helium and tritium on the same sample, and since the irradiation of the block was very non-uniform, the results have been inter-related by measurement, in a counting system of reproducible geometry, of the gross radioactivity of the spallation products in the sample.

From that part of the steel on which the cyclotron beam had been directly incident we find (from pairs of concordant measurements) ^{4}He : ^{3}He : ^{3}H : : 1·00 : 0·048 : 0·078 ; from another part, which was probably irradiated mainly by scattered particles (of rather lower energy) somewhat smaller ratios were found : ^{4}He : ^{3}He : ^{3}H : : 1·00 : 0·028 : 0·038. Correction has been made in each case for decay of ^{3}H in the interval between irradiation and measurement.

The correlation of these results with the predictions of nuclear evaporation theory (Le Couteur 1950, 1952) is rendered difficult by the fact that the theoretical results are necessarily expressed in terms of the energy of excitation of the nuclei. Protons of 340 MeV are sufficiently energetic to excite meson production, and 'knocked-on' nucleons may also be emitted before the true evaporation process commences; it is thus likely that in our sample the average excitation energy was considerably less than the full 340 MeV.

We have attempted to make allowance for this uncertainty by utilizing the data of Rudstam, Stevenson and Folger (1952) on the yields of spallation products from iron bombarded with 340 MeV protons. From their yields (with appropriate interpolation where necessary) one can deduce cross sections for events leading to the evaporation of various numbers of nucleons. Le Couteur (1950) has given graphical representations of the average yields of various evaporation products, as functions of the energy of excitation of the nucleus. From his curves, one can deduce relationships between the total number of nucleons lost by evaporation in a given event, and the average numbers of particles of various sorts constituting this total. Combining this with the cross section data of Rudstam *et al.*, one can then arrive at overall weighted average yields for the types of particle with which we are concerned. In this way we have deduced ^{4}He : ^{3}He : ^{3}H : : 1·00 : 0·068 : 0·178. In view of the assumptions involved, the agreement with our experimental results is not unsatisfactory. Our experimental value for ^{3}H : ^{4}He is rather low compared to the calculated value ; this may arise from the fact that Le Couteur's calculations (which refer specifically to a hypothetical atom of mass 100, excited so as to produce at least seven charged evaporation particles) have here been extrapolated to cover an iron nucleus ($A=56$) at moderate levels of excitation. It is to be expected that the change from $A=100$ to $A=56$, with the consequent reduction in potential barrier, would tend to reduce the yield of singly charged particles relative to doubly charged fragments.

Our thanks are due to Professor F. A. Paneth, F.R.S., for his interest, to Professor S. F. Singer (University of Maryland) who suggested the experiment, to Professor W. H. Barkas (University of California Radiation Laboratory) who kindly undertook the bombardment of the steel in the synchro-cyclotron, and to Messrs. Imperial Chemical Industries, Ltd. who have generously made available to us some of the equipment used in this investigation.

One of us (K. I. M.) is indebted to the Pressed Steel Company, Ltd., for financial support ; another (G. W.) wishes to acknowledge the receipt of an award from the Ministry of Education, during the tenure of which this work was carried out.

REFERENCES

ALLEN, M. B., and RUBEN, S., 1942, *J. Amer. Chem. Soc.*, **64**, 948.
CHACKETT, K. F., REASBECK, P., and WILSON, E. J., 1953, *Geochim. et Cosmochim. Acta*, **3**, 261.
GLÜCKAUF, E., 1946, *Proc. Roy. Soc.* A, **185**, 98.
LE COUTEUR, K. J., 1950, *Proc. Phys. Soc.* A, **63**, 259 ; 1952, *Ibid.*, **65**, 718.
LIBBY, W. F., and BARTER, C. A., 1942, *J. Chem. Phys.*, **10**, 184.
MAYNE, K. I., 1954, to be published.
RUDSTAM, G., STEVENSON, P. C., and FOLGER, R. L., 1952, *Phys. Rev.*, **87**, 358.

L. *Some Observations on the Decay of Neutral V-Particles in Photographic Emulsions**

By M. Yasin
H. H. Wills Physical Laboratory, University of Bristol†

[Received February 4, 1954]

Summary

In an examination of two-particle events recorded in photographic emulsions, four examples have been observed of a type similar to the decay of Λ^0-particles in Wilson chambers. In three of them, the measured energy-release in the decay is ∼40 mev; in the fourth example it is ∼70 mev. An event has also been observed, which appears to correspond to the decay in flight of a θ^0-particle, emitted from a nuclear disintegration of type 10+7p. One of the secondary particles arising from the decay is a negative π-meson. The measured Q value is 202 ± 11 mev.

§ 1. The Energy Release in the Decay of Λ^0-Particles

It is now well known, from experiments with Wilson chambers, that a two body decay exists of the type:—

$$\Lambda^0 \rightarrow \mathrm{p} + \pi^- + Q.$$

The available evidence has been summarized by Rochester and Butler (1953), and some of the more recent measurements are contained in the Bagnères Report (1953). Values of Q ranging from 34 mev to 45 mev have been reported by various investigators. The Pasadena group (Leighton *et al.* 1953) suggest that a second Q value of ∼75 mev also exists. We shall here describe some determinations of the energy release Q in decays of the above type observed in photographic emulsions.

In this laboratory, observers have been recording two-prong events, collectively referred to as 2+0 stars. These events belong to one of the following categories:

(*a*) elastic or inelastic scattering suffered by a charged particle;
(*b*) neutron induced nuclear disintegrations resulting in the emission of two charged particles;
(*c*) one-prong stars produced by a charged particle; and
(*d*) decays of various types of V-particles.

---

* The nomenclature used in this paper is that suggested by Amaldi *et al.* (1954).

† Communicated by Professor C. F. Powell, F.R.S.

In order to isolate events of type (*d*) from amongst the various 2+0 events recorded, the following two criteria were used :

(*a*) Only those 2+0 events were selected in which it was possible to show, by combined measurements of grain-density and multiple scattering, that one of the tracks was due to an outgoing L-meson ;

(*b*) no events were accepted in which there was a recoil track or spur of length $\geqslant 0{\cdot}5\,\mu$, or an associated electron or δ-ray at the apex of the fork. Amongst the large number of 2-prong events examined, 4 were found which satisfied the above criteria. Details of these events are given in table 1.

Table 1

Event	Exposure	Track length in microns	g^*	$p\beta$	Identity	P	E	Φ	Q
1	CM	(*a*) 800	2·23±0·074	60±19	L-meson	102±19	33±11	124°	53±13
		(*b*) 1900	3·28±0·068	101±17	Proton	321±30	54±10		
2	CM	(*a*) 1000	2·73±0·066	54±12	L-meson	96±13	30± 7		
		(*b*) 1400	3·88±0·083	95±19	Proton	304±32	49±10	127°	43±11
3	HA34	(*a*) 1800	3·15±0·063	25± 6	L-meson	62± 7	13± 3		
		(*b*) 300	3·8 ±0·128		Proton			137°	∼35
4	CS	(*a*) 4880	3·2 ±0·07		π^--meson	68± 1	16± 0·5		
		(*b*) 2800	1·98±0·05	433±87	Proton	712±88	242±53	135°	80±10

CM : Ilford G5 emulsions, 600 μ thick, exposed for 50 h, between 30 000 and 42 000 ft., with 14 cm of iron on the top and 9 cm of iron on all sides, in a Comet jet airliner flying between London and Singapore.

HA34 : Ilford G5 emulsions, 400 μ thick, exposed for 5 h at ∼65 000 ft., under 10 cm of lead.

CS : A block of Ilford G5 stripped emulsions, each 600 μ thick, exposed for 25 h between 30 000 and 42 000 ft. in a Comet jet airliner flying between London and Singapore.

Values of g^* given in col. 4 are the normalized grain-densities. Tracks of high energy electron pairs were used for the normalization and for determining the variation of development with depth in the emulsion. In event 4, track (*a*) is due to a negative π-meson, which after traversing 4·88 mm in the emulsion was brought to rest and produced a 2-prong star. In the other examples, it cannot be stated whether the L-meson is a π- or a μ-meson ; nor can the sign of the charge of the meson be determined. The identities given in col. 6 are based on the grain-density and scattering measurements. In event 3, no scattering measurements could be carried out on track (*b*). It has been attributed to a proton, however, since it could be seen visually, that for a measured value of $g^*=3{\cdot}8$, the track was not sufficiently scattered for it to have been produced by an L-meson. The space angle Φ between the two tracks in each of the examples is given in col. 9. In all the events the measurements favour the assumption that the L-meson is travelling away from the event, but in some the evidence is not decisive. In the case of event 3, however, it can be stated unambiguously that track (*a*) was produced by an outgoing π-meson. The Q values given in col. 10 have been calculated on the assumption that the L-meson is a π-meson.

The values of the momentum, p, and energy, E, given in cols. 6 and 7 of table 1 are based on scattering measurements. It is obvious, however, that such measurements on limited lengths of track, are subject to large statistical fluctuations. In order to determine the Q values more accurately, an alternative procedure was therefore employed. In each case, it was assumed that track (a) was due to a π-meson, and track (b) to a proton. Then, using the values of g^* given in col. 3 of table 1, values of $p\beta$ were deduced from the best g^* *vs.* $p\beta$ curves which had been obtained experimentally for protons and π-mesons in these emulsions. The values of p, E and Q, derived from these values of $p\beta$ are shown in table 2. Photomicrographs of events 1, 2 and 3 are shown in Plate 10.

Table 2

Event	Particle	p	E	Φ	Q*
1 (a)	π-meson	88 ± 5	25 ± 3	124°	43 ± 4
(b)	Proton	338 ± 11	60 ± 4		
2 (a)	π-meson	80 ± 4	22 ± 2	127°	41 ± 2
(b)	Proton	260 ± 5	35 ± 2		
3 (a)	π-meson	68 ± 3	16 ± 2	137°	37 ± 4
(b)	Proton	285 ± 28	47 ± 8		
4 (a)	π-meson	68 ± 1	$16 \pm 0{\cdot}5$	135°	70 ± 7
(b)	Proton	687 ± 20	226 ± 12		

* The errors shown are standard deviations; they are statistical errors associated with the grain density and angle measurements.

Since the direction of motion of track (b) in each event could not be determined unambiguously, we have also considered the following possibility. The events were assumed to represent the decay in flight of charged hyperons, the decay involving the emission of a charged L-meson, and a neutron. The velocity of the hyperon at the point of decay was obtained from the grain density of track (b) of each event. The Q values obtained from the four events for a decay of the above type, were: (1) 23 Mev, (2) 17 Mev, (3) 11 Mev and (4) 23 Mev. These values are inconsistent with other determinations of the energy release in the decay of charged hyperons.

A summary of the data presented in this section was given by the author at the International Conference on the Cosmic Radiation held at Bagnères-de-Bigorre (July 1953). At the Conference, a fifth event had also been reported, in which both the secondaries of the V-event came to rest in the emulsion and were identified as a proton and a π^--meson respectively; the Q value obtained in this example was $\sim$10 Mev. A closer examination of the event suggests, however, that it represents a nuclear interaction and not the decay of a Λ^0-particle. In view of this, we have excluded it from our statistics.

§ 2. Decay of a θ^0-Particle

A star of type 10+7p was found recently in an emulsion block consisting of 46, 600 μ thick, Ilford G5 stripped emulsions exposed at 75 000 ft. for $\sim$4 hours in England. At a distance of $\sim$100 μ from this disintegration, a two-particle event was found (see Plate 11), which appears to represent the decay in flight of a θ^0-particle. The details of this V-event are as follows :

(*a*) One of the emerging particles was a negative π-meson, for it came to the end of its range in the emulsion after travelling a distance of 3·1 mm and produced a one-prong σ-star.

(*b*) The other track has a total length of 54 mm in the stack and has been observed in eleven emulsions ; the minimum length in any single emulsion was 4 mm. The mean blob-density on the track was determined to be $b^*=0{\cdot}928\pm0{\cdot}019$ and multiple scattering measurements yielded a value of $p\beta=412\pm21$ MEV/c. Individual measurements, carried out on each of the sections of the track in the eleven emulsions, were found to be consistent with one another. From these values it may be seen that the track must have been produced by an L-meson. It is not possible to determine whether the L-meson involved is a π- or a μ-particle, nor can its direction of motion be established.

The simplest explanation would be that the V-event represents the inelastic scattering of a negative π-meson. This possibility appears to be very unlikely, however, on the following grounds :

(*a*) There is no sign of any recoil or electron track at the apex of the fork. If the event were due to an inelastic scattering, the π-meson has been degraded in energy from 340 MEV to 12 MEV in the process. That such an energy loss should take place without any visible trace, appears to be highly improbable.

(*b*) The two tracks of the V-event and the centre of the disintegration 10+7p are co-planar to within $\sim 2°$.

In view of these features, we suggest that the event is an example of the decay in flight of a θ^0-particle, the latter probably having been emitted from the star 10+7p.

The event has been analysed on the basis of this assumption, and the relevant details of the measurements are given below.

Angle between secondary mesons : $\Phi=146°,\ 39'\pm30'$

Angle Φ_- between π^--meson and assumed direction of $\theta^0=142°,\ 2'\pm30'$

Angle Φ_+ between fast light meson and assumed direction of $\theta^0=\ 4°,\ 54'\pm15'$

(It is assumed that the direction of the θ^0-particle is given by the line joining the apex of the V-event and the centre of the disintegration 10+7p.)

Range of negative π-meson	$=3128\,\mu$
Energy of negative π-meson	$=12{\cdot}2\pm0{\cdot}2$ MEV
Momentum of negative π-meson	$=59{\cdot}6\pm0{\cdot}41$ MEV/c
Transverse momentum of negative π-meson	$=37\ \ \pm0{\cdot}5$ MEV/c

	If the fast particle is a π-meson	If the fast particle is a μ-meson
Energy	342 ± 26 MeV	358 ± 27 MeV
Momentum	461 ± 27 MeV/c	452 ± 28 MeV/c
Transverse momentum	39 ± 4 MeV/c	$38{\cdot}5 \pm 4$ MeV/c
Q value	202 ± 6 MeV	220 ± 6 MeV
Mass of θ^0	$950\ m_e$	$921\ m_e$

In calculating the energy of the fast particle, account has been taken of the loss of energy by ionization along the track. The error quoted for the energy of the negative π-meson is solely that due to straggling.

The energy release Q involved in the decay

$$\theta^0 \rightarrow \pi^+ + \pi^- + Q$$

is thus determined to be 202 ± 11 MeV. This value may be compared with that of 214 MeV quoted by Thompson *et al.* (1953).

The error of ± 6 MeV has been increased to ± 11 MeV to allow for uncertainties ($\sim 5\%$) in the value of the scattering constant employed for the estimation of $p\beta$, and uncertainties in the thickness of the emulsions.

ACKNOWLEDGMENTS

I have pleasure in thanking Professor C. F. Powell, F.R.S., for his continued interest and encouragement. I am grateful to Mr. C. J. Waddington and to Mr. P. H. Fowler for drawing my attention to event 4 (§ 1) and to the event of § 2 respectively. The θ^0-event was found by Miss Sylvia Lawrence in the course of general scanning.

REFERENCES

AMALDI, E., ANDERSON, C. D., BLACKETT, P. M. S., FRETTER, W. B., LEPRINCE-RINGUET, L., PETERS, B., POWELL, C. F., ROCHESTER, G. D., ROSSI, B., and THOMPSON, R. W., 1954, *Nature, Lond.*, **173,** 123.

LEIGHTON, R. B., WANLASS, S. D., and ANDERSON, C. D., 1953, *Phys. Rev.*, **89,** 148.

ROCHESTER, G. D., and BUTLER, C. C., 1953, *Rep. Prog. Phys.*, **16,** 364.

THOMPSON, R. W., BUSKIRK, A. V., COHN, H. O., KARZMARK, C. J., and REDIKER, R. H., 1953, *Report of the Cosmic Ray Conference, Bagnères-de-Bigorre*, page 30.

LI. *Observations on Charged Hyperons in Photographic Emulsions**

By M. W. Friedlander
H. H. Wills Physical Laboratory, University of Bristol†

[Received February 3, 1954]

Summary

Two examples of charged hyperons have been observed in photographic emulsions. One decays in flight, and its transformation may be represented $Y^{\pm} \rightarrow L^{\pm}+N^0+Q$, the release of energy, Q being consistent with values obtained in other laboratories. In the other example, the secondary is a fast proton. This event may represent either the decay of a new type of hyperon or the nuclear capture of a negative hyperon.

---

§ 1. Introduction

At the Royal Society Conference in January 1953, the first decisive evidence from experiments with photographic emulsions for the existence of a charged hyperon was presented (Bonetti *et al.* 1953 a). At the same time, and by independent observations with Wilson expansion chambers, York *et al.* (1953) obtained similar evidence. These workers observed the decay in flight of an unstable particle produced in a nuclear disintegration, where the secondary particle had a mass not significantly different from that of the proton. Several examples observed in photographic emulsions and expansion chambers have since been reported from other laboratories. In some cases the hyperon is observed to decay in flight whilst in others it reaches the end of its range. The limited evidence hitherto available has been consistent with the view that we are dealing with the decay of particles of a single type, particles of which the transformations may be represented

$$Y^{\pm} \rightarrow n+\pi^{\pm}+Q \qquad (a)$$

and

$$Y^{+} \rightarrow p+\pi^{0}+Q. \qquad (b)$$

For those examples observed in photographic emulsions, only one can be interpreted in terms of (*b*) ;‡ all the others may be represented by (*a*). The Q-values for the two modes of decay appear to be equal within the present wide limits of error, $Q \sim 110$–130 MeV, and the assumed two-body decay rests upon this evidence (Bonetti *et al.* 1953 b).

---

* The nomenclature used in this paper follows the system set out by Amaldi *et al.* (1954).

† Communicated by Professor C. F. Powell, F.R.S.

‡ *Note added in proof.*—A recent event has been observed by Danysz (private communication), in which a charged hyperon, emitted from a nuclear disintegration, decays in flight, the charged secondary particle being identified as a proton. The calculated Q-value for the decay, according to (*b*), was $Q=120^{+50}_{-30}$ MeV.

The cascade decays observed in expansion chambers (Armenteros *et al.* 1952, Anderson *et al.* 1953) may also represent the decay in flight of a charged hyperon, into a light meson and a neutral particle, either Λ^0 or θ^0, but this interpretation remains to be confirmed. In each case, it depends on an observed coplanarity of one V event of a pair and the apex of the other. Whilst strongly suggestive, these observations do not prove that the neutral particle results from the decay of the charged particle. They might both result from a nuclear interaction in the matter above the chamber—a form of pair production. (See for example Fowler *et al.* 1953 b.)

In all examples of this type, reported hitherto, only lower limits can be placed on the mass of the primary particle. Of the primary particles of such events, observed in magnetic fields, all are negatively charged.

Two examples of slow charged hyperons (Y(B)1, Y(B)2) have recently been observed in stripped emulsions in this laboratory. In event Y(B)1, shown in Plate 12, a charged hyperon appears to decay in flight. The secondary, although steeply dipping, can be traced through eleven emulsions and can be identified as a light meson. In the other event, shown in Plate 13, the hyperon comes to rest. The secondary particle in this event is also steeply dipping, but can be traced through sixteen emulsions and can be identified as a proton.

These events are described in §§ 2 and 3 and are discussed in relation to other charged hyperons in § 4.

§ 2. EVENT Y(B)1

A photomicrograph of this event is shown in Plate 12. The hyperon is ejected from a star of type 26+14p and travels 4·72 mm in one emulsion before decaying in flight. All other tracks from the star have been traced either to rest, or until they left the emulsion stack. All the 26 heavily ionizing and 10 of the shower particles have been identified as protons, deuterons, α-particles or π-mesons. The remaining 4 fast particles could not be identified. The residual range of the hyperon at the instant of decay, its mean velocity during flight, and its mass have been determined by observations on the variation of gap-density and scattering along the track. These results are summarized in table 1.

Table 1. Measurements on the Primary of Y(B)1

Method	Mass of Primary m_e	Residual range at decay	$\beta = v/c$
Scattering :	2810±600	6·3±1·0 mm	0·254±0·010
Gap-density :	2670±600	6·5±1·0 mm	0·265±0·010

As a basis for further calculation, a mass value of 2750 m_e and a velocity $\beta = 0{\cdot}26$ have been assumed. The results thus obtained are not very sensitive to small changes in either of these quantities. The proper time of flight of the primary was thus found to be $5{\cdot}8 \times 10^{-11}$ sec.

The secondary is directed backwards, in the laboratory system, at a true angle of 34·5° to the direction of motion of the primary. Because the track of the secondary is very steeply inclined to the plane of the emulsion, an accurate estimate of the energy cannot be obtained from scattering measurements. The track can be traced through eleven emulsion strips until it leaves the stack, and the mean angle of scattering over the entire length is $\bar{\alpha}=0{\cdot}34^\circ/100\,\mu$. Blob counts are more reliable, and the mean normalized blob density in the eleven strips is $b^*=1{\cdot}34\pm0{\cdot}02$. This value and the observed $\bar{\alpha}$, indicates that the secondary particle was almost certainly a light meson. From the value of the blob density, and the relation between b^* and $p\beta$ already established for these plates, values of $p\beta$, assuming the secondary to be either a π- or μ-meson, can be deduced. These values are $p\beta=87\pm3$ Mev/c for a π-meson; and $p\beta=66\pm2$ Mev/c for a μ-meson. The observed energy of the meson allows us to exlude the possibility that the event represents a nuclear interaction involving, for example, a slow deuteron, for the parent particle has not sufficient kinetic energy.

Assuming that the event represents the decay in flight of a charged hyperon, and that the transformation is into two particles only, the decay may be represented

$$\mathrm{Y}^{\pm}\rightarrow\mathrm{L}^{\pm}+\mathrm{N}^{0}+Q,$$

where $\mathrm{L}^{\pm}$ denotes a light meson, and N^0 a neutral particle. Q-values and mass values for the hyperon, have been calculated for all possible combinations, assuming L to be a π- or μ-meson, and N^0 to be one of the following neutral particles already known: Λ^0 (mass 2180), n (mass 1838), B^0 (mass ~ 1400, Fowler *et al.* 1953 a), θ^0 (mass 970) and π^0 (mass 263). These values of Q and M_{Y} are set out in table 2. If the hyperon is assumed to be an excited nucleon, and if it is accepted that nucleons are conserved in these processes, then the possibilities that $\mathrm{N}^0=\mathrm{B}^0$, θ^0 or π^0 may be immediately excluded. However, if the hyperon is a created particle, then all possibilities must be allowed.

Table 2

(*a*) Q-values (Mev)

Secondary \ Neutral Particle	Λ^0	n	B^0	θ^0	π^0
π	98	100	106	115	172
μ	72	73	76	81	119

(*b*) Mass Values of Primary (m_e)

Secondary \ Neutral Particle	Λ^0	n	B^0	θ^0	π^0
π	2645	2306	1883	1468	873
μ	2528	2187	1756	1336	703

From these figures and the direct mass-measurements on the primary, it seems very unlikely that the neutral secondary is less massive than the proton. The most probable Q-value is thus ~ 100 Mev. It is, however, not possible, with the present information, to decide whether the neutral particle is a neutron or a neutral hyperon. If the latter possibility is the correct one then the event could represent a process similar to the 'cascade decays'.

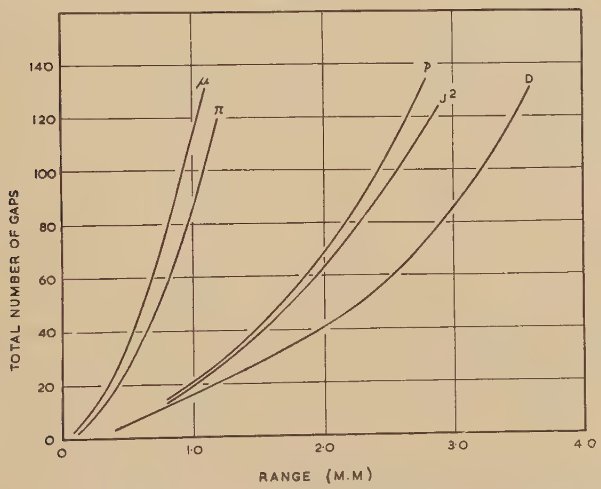
§ 3. Event Y(B)2

In this event (Pl. 13), the hyperon was ejected from a star of type 31+3n and came to rest after a path of 4·64 mm in three emulsions. All other tracks except the shower particles, have been followed from the star and identified as protons, deuterons, α-particles, or π-mesons. From the end of the hyperon track, a second charged particle emerges, with a track steeply inclined to the plane of the emulsion, which leaves the stack after 13·2 mm, having passed through sixteen emulsions. The measurements on the primary are summarized in table 3.

Table 3. Mass Measurements on Primary of Y(B)2

Method :	Range-scattering	Grain-density-range
Mass :	$2680 \pm 350\ m_e$	$2200 \pm 350\ m_e$

Fig. 1



Total number of gaps *vs.* range, from the end of the track. Comparison curves are given for deuterons, protons and π- and μ-mesons; the curve for the hyperon is that denoted by J2.

In making the range-scattering measurements, three different sets of cell sizes were employed in conjunction with the constant sagitta method (Biswas *et al.* 1953). The value quoted is the mean of the three determinations. Gap measurements all indicate a mass between that of the proton and the deuteron, but the experimental conditions render the observations unreliable and it is therefore not possible to give a definite mass value. A plot of the total number of gaps as a function of range, is shown in fig. 1, which includes curves for μ- and π-meson, protons and deuterons, for comparison.

As in event Y(B)1, the track of the secondary particle is very steep. In the first three plates counting from that in which the hyperon comes to rest, the secondary has a mean grain-density of 29·6 grains/50 μ.

Scattering measurements give a value of $\bar{\alpha}=0{\cdot}25°/100\,\mu$. These measurements show that the secondary particle was considerably heavier than the π-meson ; and taking account of the fact that the value of $\bar{\alpha}$ is an upper limit, the track being steeply dipping, they are consistent with the assumption that it was a proton. If the secondary particle were not a proton, but a lighter particle, then in the length observed in the emulsions (13·2 mm) a significant change of grain density should be observed. No such change has been found. The grain density and gap measurements have therefore been employed to estimate the total range of the proton, and a value $R=3{\cdot}0\pm0{\cdot}8$ cm has thus been obtained. This corresponds to a proton energy of 100±15 Mev.

We may consider two possible explanations for this event. If it represents the decay of a charged hyperon

$$\mathrm{Y}^+\rightarrow \mathrm{p}+\pi^0+Q,$$

then it is easily shown that the energy release, Q, is of the order of 430 Mev, a value considerably higher than any previously reported for any similar event interpreted as due to the decay of a charged hyperon.

An alternative explanation is that the event represents the nuclear capture of a negative hyperon, the proton being the only visible product of the disintegration.

§ 4. Discussion

A recent paper by Bonetti *et al.* (1953 b) summarizes the results available until the beginning of November. Since then two hyperons have been observed by King *et al.* (1953) ; one of these hyperons comes to rest and has a fast π-meson secondary. Considering two possibilities for the neutral secondary for a two body decay, viz. Λ^0 and a neutron, they obtain Q values of 101 and 103 Mev. Event Y(B)1 is in good agreement with this. The events listed by Bonetti *et al.* have slightly higher Q values, but within the accuracy of all the measurements, limited by steepness or short track length of the secondaries, all of the decays so far observed are not mutually inconsistent. In appearance event Y(B)1 is very similar to that observed in Padua (Ceccarelli and Merlin 1953), which event has a slightly larger Q value.

Observations on cascade decays may be taken as supporting the identification of the neutral secondary, in the charged hyperon decay, as a Λ^0, but the weight of these results is not yet very large.* In this case, the Q-values would be reduced by $\sim$2 MEV as compared with the case for a neutron secondary. The track length of the primary particles is in no case sufficiently long to distinguish between the different hyperon masses corresponding to these two possible identities of the neutral particle.

---

* *Note added in proof.*—At the meeting of the American Physical Society in New York (January, 1954), Cowan presented the results of a re-examination of a cascade decay (*Bagneres Report*, p. 99). In this event, the neutral V-particle is identified as a Λ^0, and the plane of the neutral V-particle contains the decay point of the charged V-particle. Assuming this to represent a decay of the form $\mathrm{Y}^-\rightarrow\Lambda^0+\pi^-+Q$, the Q-value is calculated to be ~65 Mev. In addition, a re-examination (Butler, private communication) of the first cascade decay (Armenteros *et al.* 1952), on the basis of the decay mentioned above, shows a Q-value ~15–50 Mev, possibly consistent with the event of Cowan. Event Y(B)1 does not therefore appear to be of this cascade type.

Event Y(B)2 does not fit into either of the decay schemes discussed above. If the possibility of a three-body decay be excluded, then this event represents either a decay involving a very high energy release, or a nuclear capture with subsequent disintegration. The latter would seem the most probable explanation, without necessitating the introduction of a new type of hyperon. Since the events described above were observed at Bristol, Johnston and O'Ceallaigh (1954) have also observed an event which seems to represent the nuclear capture of a negative hyperon. This event is described in detail in the following paper.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge, with sincere thanks, the assistance received from Mr. P. H. Fowler in some of the measurements, and from him and Dr. M. G. K. Menon and Dr. D. H. Perkins in many discussions ; also Professor C. F. Powell for his constant encouragement and interest, and Miss P. Dyer and Mr. A. R. Gattiker for the photographs, and Miss J. Witchell for the diagram. The events described in this paper were observed by Miss R. Mitchell and Miss A. Stradling, in the course of general scanning.

REFERENCES

AMALDI, E., ANDERSON, C. D., BLACKETT, P. M. S., FRETTER, W. B., LEPRINCE-RINGUET, L., PETERS, B., POWELL, C. F., ROCHESTER, G. D., ROSSI, B., and THOMSON, R. W., 1954, *Nature, Lond.*, **173**, 123.

ANDERSON, C. D., COWAN, E. W., LEIGHTON, R. B., and VAN LINT, V. A. J., 1953, *Bull. Amer. Phys. Soc.*, **28**, (no. 5) 17.

ARMENTEROS, R., BARKER, K. H., BUTLER, C. C., CACHON, A., and YORK, C. M., 1952, *Phil. Mag.*, **43**, 597.

BISWAS, S., GEORGE, E. C., and PETERS, B., 1953, *Proc. Ind. Acad. Sci.*, **38**, 418.

BONETTI, A., LEVI-SETTI, R., PANETTI, M., and TOMASINI, G., 1953 a, *Nuovo Cim.*, **10**, 345 ; 1953 b, *Ibid.*, **10**, 1736.

CECCARELLI, M., and MERLIN, M., 1953, *Nuovo Cim.*, **10**, 1207.

FOWLER, W. B., SHUTT, R. P., THORNDIKE, A. M., and WHITTEMORE, W. L., 1953 a, *Phys. Rev.*, **91**, 1287 ; 1953 b, *Phys. Rev.* (in the press).

JOHNSTON, R. H. W., and O'CEALLAIGH, C., 1954, *Phil. Mag.*, **45**, 424.

KING, D. T., SEEMAN, N., and SHAPIRO, M. M., 1953, *Phys. Rev.*, **92**, 838.

REPORT OF THE COSMIC RAY CONFERENCE, Bagneres-de-Bigorre, 1953.

YORK, C. M., LEIGHTON, R. B., and BJORNERUD, E. K., 1953, *Phys. Rev.*, **90**, 167.

LII. *Evidence for Nuclear Interaction of a Charged Hyperon Arrested in Photographic Emulsion*

By R. H. W. JOHNSTON and C. O'CEALLAIGH
School of Cosmic Physics, Dublin Institute for Advanced Studies†

[Received February 3, 1954]

SUMMARY

An event which is interpreted as a nuclear interaction of a charged hyperon has been observed in a photographic emulsion. The appearance of the interaction is that of a typical σ-star, but is caused by a particle of protonic mass. Estimates of mass by two independent techniques are in agreement with each other, and are consistent with those of charged hyperons reported by other workers.

§ 1. INTRODUCTION

DURING examination of plates from a stack of $600\,\mu$ stripped emulsions exposed during the Sardinian Expedition of Summer 1953, an event has been found which presents the following unusual features.

A particle of $g^*{\sim}7$ is emitted from a star of 30 branches of which the majority are 'black' or 'grey'. Because the star is very close to the surface of the emulsion, it has been considered unprofitable, for the moment, to undertake a more precise classification of this interaction. There are also other reasons which are connected with the processing of the plates. These will be discussed in the appendix. It is clear, however, that a heavy nucleus is involved, and, further, the energy per track is of order 100 MeV.

Thus, the creation of hyperons (Fretter *et al.* 1953) appears energetically possible. The particle under consideration which resembles a proton, traverses 3 emulsions, and stops after a range of $7350\,\mu$. That the particle was moving with negligible speed at the end of its track, was established by observing the distribution of gaps; this distribution was compared with that of protons brought to rest in the same region of the emulsion. A photomicrograph of the event is shown in Plate 14. From the point of arrest two particles are emitted. One of these is of very short range $\sim 4\,\mu$. It appears to lie in a plane parallel to the surface of the emulsion. No charged particle can be observed in association with its end. The other particle is emitted at a very steep angle. It traverses three complete emulsions, but cannot be traced into the fourth. It is possible therefore that it came to rest very close to the glass backing plate, or in the paper tissue separating the emulsion strips. Since the end could not be scrutinized, we cannot exclude the possibility of subsequent interaction or decay. Its appearance, however, is that of a singly-charged particle more massive than a π-meson, and it seems reasonable to suppose that it is a proton. The residual range cannot be measured with high accuracy, but it is approximately 2000 μ (proton energy ~20 MeV).

---

† Communicated by the Authors.

Because of the following observations, it is clear that the event cannot be interpreted as a case of elastic scattering of a proton of residual range 2000 μ, with recoil of range 4 μ. The tracks are not coplanar, and momentum cannot be conserved unless we invoke the emission of at least one neutral particle. Moreover, as has been remarked, the gap-density of the incoming track at the point of supposed scattering differs significantly from that of a proton of the postulated residual range. It is concluded therefore, that we are dealing with a nuclear interaction similar to those commonly produced by π-mesons arrested in the emulsion.

§ 2. MASS OF INTERACTING PARTICLE

Mass estimates of the interacting particle have been made using two independent methods, and these agree within the expected statistical uncertainty.

(1) *Multiple-Scattering/Residual Range*

The method of constant sagitta described by the Brussels and Bombay Groups (1953) was used with the following modifications. An estimate of noise was made by the method of cross products (Menon *et al.* 1951, Molière 1951). Large unrepresentative signals were treated by the substitution method of Goldsack (1953). The range increment tables $P=1$ and $\pi=1\cdot6$ kindly supplied by Dr. Peters were used as a basis for these measurements. Evidence for the presence of distortion was found from the distribution of the signals and the values of the cross products. The necessary correction was small, and was applied by the Cosyns–Vanderhaege method using measurements on neighbouring steeply dipping tracks. A small correction was also made for the influence of dip. The corrected mass-estimates were:

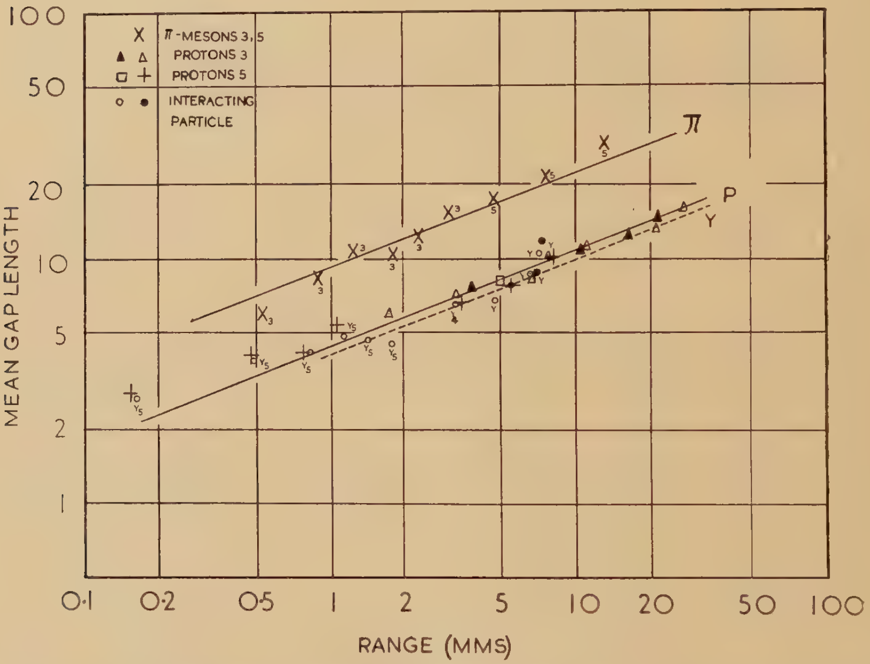
$$P=1\cdot0 \qquad 2280\,{}^{+700}_{-480}\, m_e$$

$$\pi=1\cdot6 \qquad 2100\,{}^{+600}_{-400}\, m_e$$

(2) *Mean Gap-Length/Residual Range*

The conditions of processing the plates which were cleared after fixing, were such that the plateau blob-density was low—14 blobs/100 μ. In consequence, the blob-density/range characteristics were unsatisfactory. For protons of residual range R, $2\text{ mm}\leqslant R\leqslant 15\text{ mm}$, the blob-density/range curve passes through a flat maximum so that it appears to be independent of range. The theory of this effect has been investigated by one of us (O'Ceallaigh 1953 a) and will be discussed in a later communication. In order to estimate ionization in plates such as the present, observations must be made of the variation with range of parameters such as the photoelectric density or the mean gap-length $\bar{G}$ (O'Ceallaigh 1953 a, Menon and O'Ceallaigh 1954). As hitherto described, neither of these methods is convenient or rapid, and the method $\bar{G}/R$ was therefore modified as described in the appendix.

Estimates of $\bar{G}$ were made in the three plates through which the interacting particle passed, and have been plotted on log–log paper as a function of residual range (see figure).



Estimate of mass of interacting particle by $\bar{G}/R$.

The interacting particle was emitted from a star in plate 3, passed through 4 and came to rest in plate 5. The experimental points are numbered accordingly, except for those of the interacting particle measured in plate 3 which are labelled Y with no subscript. The plate numbers and distinguishing marks for the calibration particles are to be found on the figure. $\bar{G}$ in micrometer drum divisions, $11{\cdot}4 = 1\ \mu$.

These values of $\bar{G}$ have been compared with those found for identified π-mesons and stopping protons, of which the tracks passed through the suitable regions in the plates. There was found evidence for the presence of protons showing anomalously low ionization which may have been those of faded or ' wet ' tracks. For this reason, the calibration protons, with one exception, were chosen from amongst those which could be traced back to a nuclear interaction in the emulsion. The values obtained from individual tracks are distinguished by symbols listed on the diagram. That the selected particles were very unlikely to have been deuterons or tritons was established (*a*) by observation of the associated δ-rays, (*b*) by measurement of blob-density in regions beyond the extremum, and finally (*c*) by mass estimate by range-scattering using Peters' method.

The proton points have been fitted visually by a straight line. This line will be seen to be a satisfactory fit. Except for points of small residual range where the statistical weight is low, the points are based on segments of track $\sim 1000\,\mu$, each containing some 500 visible gaps. Since we have not yet investigated the problem of the sampling errors of our estimates of $\bar{G}$, it has seemed pointless to attempt to fit by an analytical method.

The π-line has been drawn parallel to the proton line in the position calculated from the known mass-ratio, and is in good agreement with the experimental points. The open circles marked Y are the experimental points for the interacting particle. Of the latter, one point corresponding to the segment nearest the star indicated a mass value less than that of the proton. That this was probably due to its nearness to the surface of the plate, is shown by the pair of points in full circle obtained by dividing this segment in two. The point of high $\bar{G}$ lies in a region $<30\,\mu$ from the surface, and is rejected, the other agrees with the remaining observations. A hatched line, corresponding to mass 2300 m_e has been drawn parallel to the proton line, and appears to fit the points in a reasonable manner.

The position of the first 6 of these may be compared with those of a proton stopping in the same plate (upright crosses). Although the points in this region are of inferior statistical weight, the agreement between them demonstrates that we are dealing with a particle which was at rest, or moving with negligible velocity, at the point of interaction.

§ 3. DISCUSSION

The satisfactory agreement between the mass-estimate by two independent methods, makes it reasonable to suppose that the event observed is due to the absorption of a negative hyperon in accordance with some mechanism such as

$$Y^- + P = n + n + Q. \qquad \dots\dots\dots \quad (1)$$

Taking the mass of Y^- as 2300 m_e, the expected value of Q is about 230 MEV. The appearance of the interaction is quite consistent with such value of Q. It is also consistent with that expected on certain hypotheses (Friedlander *et al.* 1953), when a negative K-particle is absorbed in a nucleus. Although the interconnecting particle is not a π-meson, it cannot perhaps be excluded that it is a negative K-meson. The agreement between the independent estimates of mass is such, however, that it does not seem possible to accept this identification without assuming unreasonable values for the statistical uncertainties of the measurements.

The present event may be compared with that reported in this issue by Friedlander and called Y(B)2 and with one described by Bonetti *et al.*

(1953). The latter event J_{pl} has been interpreted as the decay of a hyperon in accordance with the scheme

$$J_{pl} \rightarrow P + \pi_0. \qquad \ldots \ldots \ldots \quad (2)$$

The proton from J_{pl} has a range of 1670±18 μ. Thus it, and that described by Friedlander, cannot both be cases of the process (2). It is possible, however, to interpret all three events as an interaction of Y^- in accordance with (1), although as pointed out by Bonetti *et al.*, owing to the short range (900 μ) of the primary particle of J_{pl}, they could not exclude the possibility that their event represents the capture of a negative K-particle.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge our indebtedness to the team of microscope observers of this School, in particular Mrs. Máirín Johnston by whom the event was found, Miss Peggy O'Hea who gave valuable assistance with the measurements, Miss P. Dyer and Mr. A. R. Gattiker for the photomicrograph. We also wish to thank Professor C. F. Powell and the other organizers and members of the 1953 Sardinia Expedition for the exposures, and the members of the H. H. Wills Laboratory by whom the plates were processed. One of us (R. H. W. J.) wishes to thank the Council of the Dublin Institute for Advanced Studies for a Research Scholarship.

APPENDIX

Measurement of Mean Gap-Length ($\bar{G}$) Improved Technique

The rapidity of the technique for estimating $\bar{G}$ described in earlier communications has been much improved by the modifications now to be described. In essence, this procedure has been suggested independently by Fowler (1953). An eyepiece is furnished with a set of four spider webs placed at suitable distances apart. A convenient choice consists of intervals of 0·5, 1·0, 2·0 and 4·0 μ all measured from the first web. Every visible gap is observed and may be classified immediately according to which intervals it exceeds. Thus, for each segment of track, we may plot on log-linear paper, five points of an integral gap-length distribution, and after a simple correction for dip, a straight line may be fitted visually to the corrected points. Alternatively, it may be shown that if N_{ϵ_1} is the number of gaps exceeding ϵ_1, and N_{ϵ_2} the number exceeding ϵ_2, then the mean gap-length $\bar{G}$ may be found from the relationship (O'Ceallaigh 1953 b)

$$\frac{N_{\epsilon_2}}{N_{\epsilon_1}} = \exp -(\epsilon_1 - \epsilon_2)/\bar{G}. \qquad \ldots \ldots \ldots \quad (3)$$

The validity of this relationship is unaffected by changes of processed grain-diameter, and the elaborate corrections for depth necessary in photoelectric techniques are thus avoided. It is necessary, however, to exclude observations made very close to the bounding surfaces of the emulsion, as conditions of development are commonly anomalous in these regions.

In practice, it will be found convenient to estimate $\bar{G}$ from the slope of the straight line fitted as described to the experimental points. The validity of the method has been tested by comparison with direct measurement of $\bar{G}$ using a filar micrometer. The agreement was satisfactory. The principal advantage of the method is its rapidity which is an improvement by a factor of about 5 on the direct measurement of $\bar{G}$ by the filar micrometer. Disadvantages are that some information is lost, and that unlike those in a direct estimate of $\bar{G}$, the sampling distributions remain to be investigated.

It is intended to examine the application of the technique to the classification of the branches of stars in circumstances, such as the present, where direct blob/gap density measurements do not yield useful information.

REFERENCES.

BONETTI, LEVI SETTI, PANETTI and TOMASINI, 1953, *Nuovo Cim.*, **10**, 1736.

BRUSSELS AND BOMBAY GROUPS, 1953 (Dilworth, Goldsack, Hirschberg, Biswas, George, Lal, Yash-Pal), *Bagnères Conference Report*, 131.

FOWLER, P. H., private communication.

FRETTER, GREGORY, JOHNSTON, LAGARRIGUE, MEYER, MULLER and PEYROU, 1953, *Bagnères Conference Report*, 26.

FRIEDLANDER, 1954, *Phil. Mag.*, **45**, 418.

FRIEDLANDER, HARRIS and MENON, 1953, *Bagnères Conference Report*, 161.

GOLDSACK, 1953, private communication.

MENON and O'CEALLAIGH, 1954, *Report of London Conference*, January 1953, *Proc. Roy. Soc.* A, **221**, 306.

MENON, O'CEALLAIGH and ROCHAT, 1951, *Phil. Mag.*, **42**, 932.

MOLIÈRE, 1951, private communication, unpublished.

O'CEALLAIGH, 1953 a, *Bagnères Conference Report*, 73; 1953 b, unpublished.

LIII. *Proton-Gamma Angular Correlations and Gamma-Ray Angular Distributions in the* ^{10}B(α, pγ)^{13}C *Reaction*

By A. G. STANLEY
Cavendish Laboratory, Cambridge*

[Received January 22, 1954]

ABSTRACT

The angular distributions of the high energy gamma-rays (a mixture of 3·7 and 3·9 MeV gamma-rays) and of the low energy gamma-rays (0·2 MeV) from the ^{10}B(α, pγ)^{13}C reaction, and the angular correlations of the high energy gamma-rays with the emitted protons have been measured at five alpha-particle resonances at 1·13, 1·51, 1·64, 1·68 and 1·83 MeV. These experiments confirm the assignment of spins and parities 4^-, 3^- and 4^+ to excited levels of ^{14}N at 12·42, 12·69 and 12·78 MeV, and spins and parities $3/2^-$ and $5/2^+$ to excited levels of ^{13}C at 3·68 and 3·89 MeV.

§ 1. INTRODUCTION

WE have made a detailed study of the ^{10}B(α, pγ)^{13}C reaction with the electrostatic generator of the Cavendish Laboratory. A previous article (Shire *et al.* 1953), hereafter referred to as I, gives an account of some of these investigations. The present paper describes experiments on the angular distribution of the high energy gamma-rays (a mixture of 3·7 and 3·9 MeV gamma-rays) and the low energy gamma-rays (0·2 MeV) from this reaction, and experiments on the angular correlation of the short-range protons with the high energy gamma-rays. We have measured the distributions and correlations for five alpha-particle resonances at 1·13, 1·51, 1·64 and 1·83 MeV. From the results we were able to confirm the spins and parities assigned in paper I to levels of ^{13}C at 3·68 and 3·89 MeV, and to the excited levels of ^{14}N corresponding to the first three alpha-particle resonances.

§ 2. EXPERIMENTAL METHOD AND APPARATUS

We made all measurements with a beam of singly charged helium ions produced by the electrostatic generator. We used thin targets of separated ^{10}B, about 20 μg cm^{-2}, on thin silver or aluminium backing. The gamma-rays were detected by means of a sodium-iodide crystal and a photomultiplier.

---

* Communicated by E. S. Shire.

2.1. *Proton-Gamma Angular Correlations and Angular Distributions of High Energy Gamma-Rays*

For the proton-gamma angular correlations we used two arrangements : in the first the gamma-ray detector could be rotated in the vertical plane perpendicular to the incident alpha-particle beam ; in the second it was rotated in the horizontal plane containing the incident beam. In both arrangements we detected the protons with a proportional counter fixed in the horizontal plane at right angles to the incident beam. In most of our experiments we turned the gamma-ray detector through an angular range from 90° to 180° relative to the proportional counter, but a few readings were taken at forward angles. The solid angles of acceptance of the proportional counter and of the sodium-iodide crystal were usually 0·12 and 0·15 steradians respectively, but we sometimes used smaller solid angles to measure the gamma-ray distributions.

We used a conventional circuit with a resolving time of 0·2 μsec to detect coincidences between the protons and the gamma-rays. In order to record true and accidental coincidences simultaneously, we delayed the pulses from one of the two channels by 2·5 μsec before passing them into a second coincidence unit together with the undelayed pulses from the other channel (Littauer 1950). The ratio of the resolving times of the two coincidence units was determined before and after each experiment ; it remained constant over a period of several months. We monitored the reaction by means of the proton counts.

In our experiments we were unable to separate the protons and gamma-rays from the following three reactions (see paper I), which we shall refer to as (A), (B) and (C) :

$$\begin{array}{lll}
\text{(A)}\quad {}^{10}\mathrm{B}+{}^{4}\mathrm{He}\rightarrow{}^{14}\mathrm{N}^{*}\rightarrow & {}^{13}\mathrm{C}^{*}\ (3{\cdot}9\ \textsc{mev}) & +\mathrm{p}_3 \\
 & \downarrow & \\
 & {}^{13}\mathrm{C}\ \ (\text{ground state}) & +\gamma(3{\cdot}9\ \textsc{mev}). \\
\text{(B)}\quad {}^{10}\mathrm{B}+{}^{4}\mathrm{He}\rightarrow{}^{14}\mathrm{N}^{*}\rightarrow & {}^{13}\mathrm{C}^{*}\ (3{\cdot}9\ \textsc{mev}) & +\mathrm{p}_3 \\
 & \downarrow & \\
 & {}^{13}\mathrm{C}^{*}\ (3{\cdot}7\ \textsc{mev}) & +\gamma(0{\cdot}2\ \textsc{mev}) \\
 & \downarrow & \\
 & {}^{13}\mathrm{C}\ \ (\text{ground state}) & +\gamma(3{\cdot}7\ \textsc{mev}). \\
\text{(C)}\quad {}^{10}\mathrm{B}+{}^{4}\mathrm{He}\rightarrow{}^{14}\mathrm{N}^{*}\rightarrow & {}^{13}\mathrm{C}^{*}\ (3{\cdot}7\ \textsc{mev}) & +\mathrm{p}_2 \\
 & \downarrow & \\
 & {}^{13}\mathrm{C}\ \ (\text{ground state}) & +\gamma(3{\cdot}7\ \textsc{mev}).
\end{array}$$

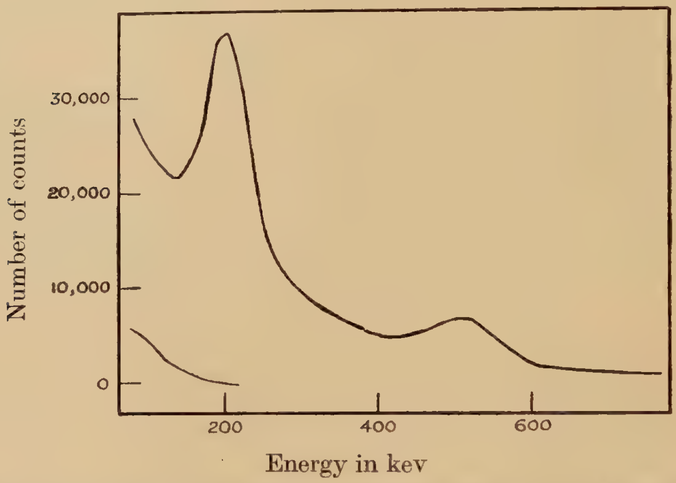
A determination of the branching ratio of the gamma-rays showed that the total yields of reactions (A) and (B) were in the ratio 7 : 3. The yield from reaction (C) was very weak except at the 1·51 Mev resonance, where this reaction contributed 16% of the total proton counts. Deuterons from the $^{10}\mathrm{B}(\alpha, \mathrm{d})^{12}\mathrm{C}$ reaction were also detected by the proportional counter, but did not produce any true coincidences.

2.2. *Angular Distributions of Low Energy Gamma-Rays*

We studied the low energy gamma-rays by means of an electronic pulse analyser of the type described by Hutchinson and Scarrott (1951).

We found that the 0·2 MeV gamma-rays due to reaction B were superimposed on a large background of low energy pulses. Some of these were produced by the generator and the remainder by high energy radiation from the target that had been scattered and degraded by the sodium-iodide crystal. We reduced the effect of the former as much as possible by surrounding the crystal with a thick lead screen, in which we cut a hole, 1 in. in diameter, opposite the target. Figure 1 shows the spectrum of gamma-rays of energy less than 700 kev after screening the crystal. The two peaks at 200 and at 500 kev were produced by gamma-rays from reaction (B) and by annihilation radiation respectively.

Fig. 1



Spectrum of gamma-rays of energy less than 700 kev at the 1·51 MeV resonance. The gamma-rays were detected at 90° to the alpha-particle beam. The lower curve shows the background radiation from the generator after screening.

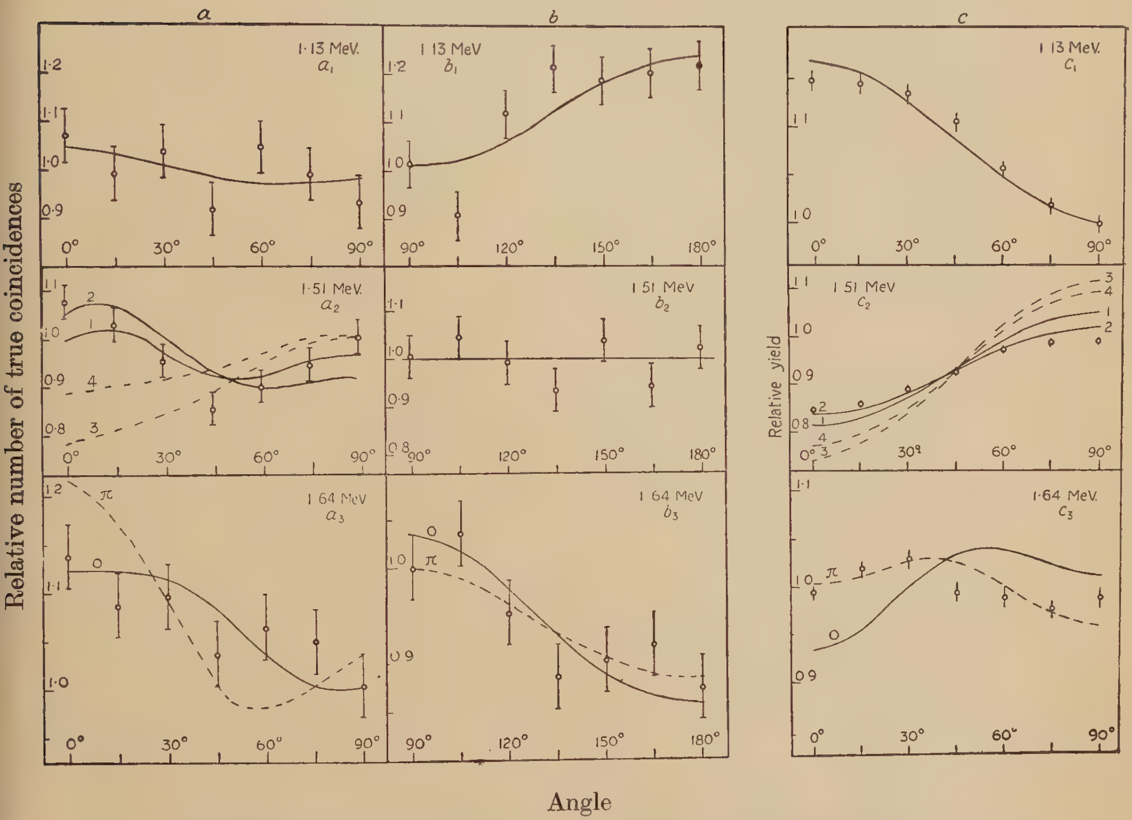
We made measurements with and without a piece of lead foil, 0·115 cm thick, placed in front of the aperture. We found that the reduction of the low energy background was proportional to the reduction of the high energy radiation measured simultaneously. From a calculation we estimated that the lead foil should have absorbed 76% of the incident 0·2 MeV radiation. From these measurements and from similar measurements with only the background due to the generator present, we were able to determine the amount of 0·2 MeV radiation relative to the amount of high energy radiation from the target.

§ 3. EXPERIMENTAL RESULTS

Figures 2, 3 and 4 show the experimental angular correlations and gamma-ray distributions after correction for the finite geometry of the detectors. We were able to compare the results for the 1·13, 1·51 and 1·64 MeV resonances with theoretical predictions (see § 4); the interference produced by overlapping excited states of ^{14}N (see paper I) made similar comparisons impossible for the resonances at 1·68 and 1·83 MeV.

The angular distributions of the 0·2 MeV gamma-rays are shown in fig. 4. The standard deviations of the experimental points are rather large for the 1·13 MeV resonance owing to the low reaction yield, and for the 1·83 MeV resonance owing to the increased background from the generator.

Fig. 2

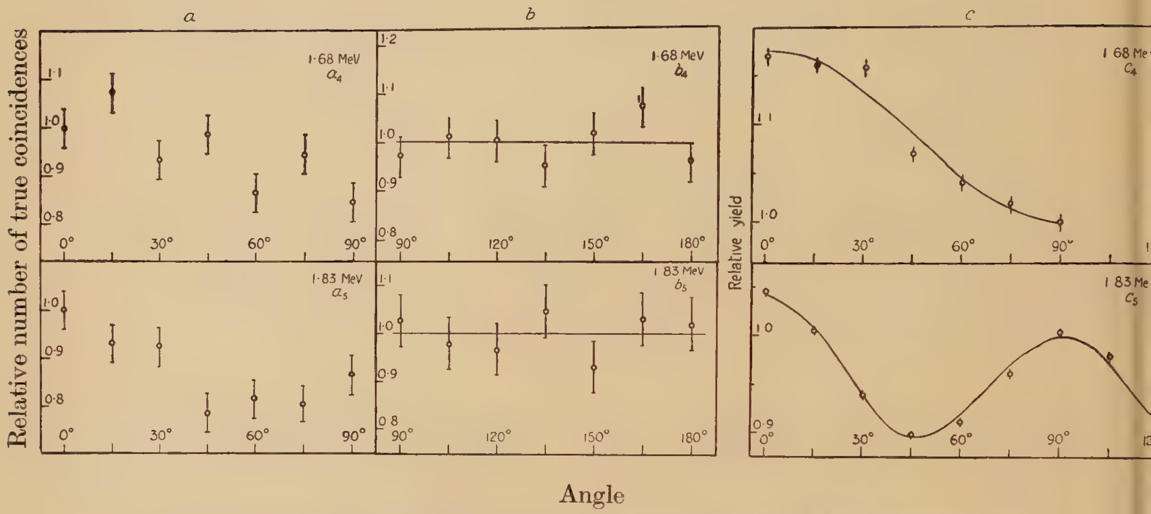


Proton-gamma angular correlations in plane of incident alpha-particle beam (column *a*), in plane perpendicular to incident beam (column *b*) and angular distributions of high energy gamma-rays (column *c*). In columns *a* and *c* the angles are measured with respect to the incident beam, in column *b* with respect to the direction of the observed protons in laboratory space. ———— – – – – – Theoretical curves. The relative phases of the spin-channel amplitudes assumed in reactions (A) and (B) for the 1·64 MeV resonance are as shown; for the 1·51 MeV resonance the relative phases assumed are :

Curve	Reactions (A) and (B)	Reaction (C)
1	0	0
2	0	π
3	π	0
4	π	π

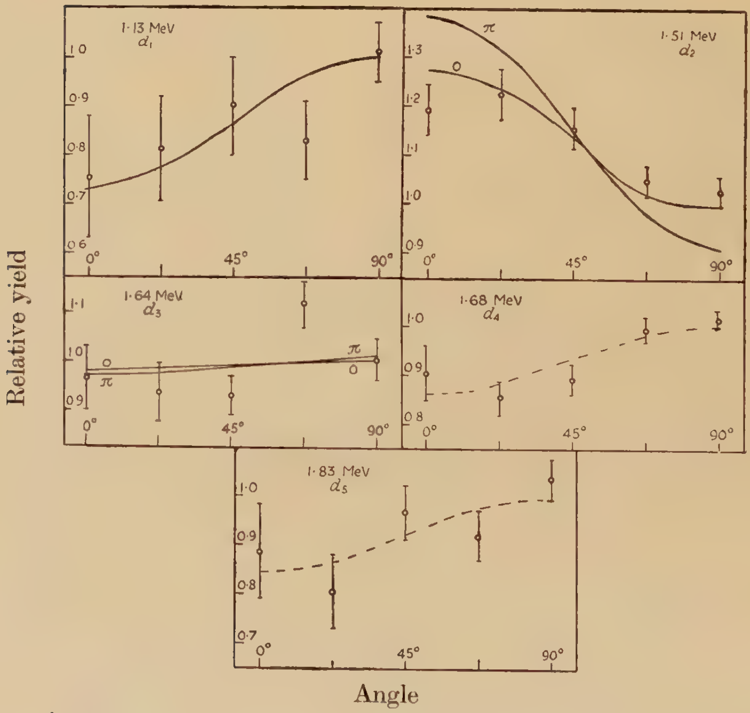
In b_2 all the theoretical curves are isotropic within experimental error. Magnetic dipole transition has been assumed for the 3·7 MeV gamma-rays in all theoretical curves.

Fig. 3



Proton-gamma angular correlations in plane of incident alpha-particle beam (column *a*), in plane perpendicular to incident beam (column *b*) and angular distributions of high energy gamma-rays (column *c*). The angles are defined as in fig. 2. The smooth curves in column *c* have been fitted to the experimental results by the method of least squares.

Fig. 4



Angular distributions of low energy (0·2 MeV) gamma-rays. ——— Theoretical curves, – – – – – – experimental curves obtained by method of least squares. The relative phases of the spin-channel amplitudes assumed for the resonances at 1·51 and 1·64 MeV are as shown.

§ 4. COMPARISON OF EXPERIMENTAL RESULTS AND THEORETICAL PREDICTIONS

4.1. *Theory*

Biedenharn *et al.* (1951) have derived a formula for three-stage angular correlations, but this does not cover the case where the emitted particle has non-zero spin. We have made use of a method, employed by Spiers (1950) for two-stage correlations, in which the angular momentum wave function for the final state of the whole system is obtained by successive transformations from the initial state. In the final state all the emitted particles are at a very large distance from the residual nucleus, so that the radial part of the wave function may be omitted from the correlation formula. Taking the direction of the incident alpha-particle beam as the axis of reference and assuming that only one level of the compound nucleus, ^{14}N, contributes to the reaction, the theoretical angular correlation between protons emitted in the direction (θ_1, ϕ_1) and gamma-rays emitted in the direction (θ_2, ϕ_2) is given by

$$\begin{aligned} W(\theta_1, \phi_1 ; \theta_2, \phi_2) = \sum_{M_0, M_1'', M_2} & \Big| \sum_{l_0} i^{l_0}(2l_0+1)^{1/2} \langle S_0 M_0 l_0 0 \,|\, JM \rangle \\ & \times \sum_{S_1, l_1} A(J, S_0, S_1, l_0, l_1) \sum_{M_1} \langle JM \,|\, S_1 M_1 l_1 m_1 \rangle \\ & \times Y_{l_1}^{m_1}(\theta_1, \phi_1) \langle S_1 M_1 \,|\, S_1' M_1' S_1'' M_1'' \rangle \\ & \times \sum_{j_2} D(S_1', M_1', S_2, M_2) A(j_2) \Phi_{j_2}^{m_2}(\theta_2, \phi_2) \Big|^2 . \quad . \quad . \quad . \quad (1) \end{aligned}$$

S_0 is the resultant spin of alpha-particle and target nucleus, ^{10}B, and is thus always 3. J is the total angular momentum of the compound nucleus, ^{14}N. The spins S_1' and S_1'' of the excited level of ^{13}C and of the proton are combined vectorially to produce the channel spin S_1. The ground level of ^{13}C has spin $S_2=\frac{1}{2}$. The symbols M denote the components in the direction of the incident beam of the spins with the corresponding suffixes.

$$\langle S_0 M_0 l_0 0 \,|\, JM \rangle, \quad \langle JM \,|\, S_1 M_1 l_1 m_1 \rangle \quad \text{and} \quad \langle S_1 M_1 \,|\, S_1' M_1' S_1'' M_1'' \rangle$$

are Clebsch–Gordon transformation coefficients, and $Y_{l_1}^{m_1}(\theta_1, \phi_1)$ is a spherical harmonic for the emitted proton. $D(S_1', M_1', S_2, M_2)$ is a transformation coefficient for the multipole field of the gamma-ray and $\Phi_{j_2}^{m_2}(\theta_2, \phi_2)$ its angular dependence (Condon and Shortley 1951).

The factor $A(J, S_0, S_1, l_0, l_1)$ is the product of the following terms :

$$A(J, S_0, S_1, l_0, l_1) = K(J, E_0) f(l_0, E_0) N(J, S_0, S_1, l_0, l_1) f(l_1, E_1). \quad . \quad . \quad . \quad . \quad . \quad . \quad (2)$$

$K(J, E_0)$ is a function of the alpha-particle energy E_0 usually expressed in the form of a complex resonance denominator. $f(l_0, E_0)$ and $f(l_1, E_1)$ are complex factors describing the probability amplitudes associated with orbital momenta, l_0 and l_1, of the incident and emitted particles. The intrinsic nuclear factor $N(J, S_0, S_1, l_0, l_1)$ depends upon the spins and orbital momenta in a wholly unknown way. The dispersion theory of Wigner and Eisenbud (1947) suggests that it is real, but it may be either positive or negative. Since the proton has spin $\frac{1}{2}$, the channel spin S_1 may in general take two values, which in turn give rise to two values of $N(J, S_0, S_1, l_0, l_1)$. In paper I the ratio of the squares of the latter has been defined as the spin-channel factor. It may be seen from eqn. (1) that the two spin channels interfere coherently, i.e. either by addition or by subtraction of the spin-channel amplitudes. The nuclear matrix element $A(j_2)$ associated with the multipole j_2 (see Lloyd 1951) is independent of the magnetic substates.

From eqn. (1) we can derive an expression for the angular distribution of the gamma-rays by integrating the spherical harmonic for the proton, which is not observed, over all space. The resulting angular distribution is given by

$$\begin{aligned} W(\theta_2)= & \sum_{M_0, M_1'', M_2} \sum_{l_1, m_1} \Big| \sum_{l_0} i^{l_0}(2l_0+1)^{1/2} \langle S_0 M_0 l_0 0 \mid JM \rangle \sum_{S_1} A(J, S_0, S_1, l_0, l_1) \\ & \times \langle JM \mid S_1 M_1 l_1 m_1 \rangle \langle S_1 M_1 \mid S_1' M_1' S_1'' M_1'' \rangle \\ & \times \sum_{j_2} D(S_1', M_1', S_2, M_2) A(j_2) \Phi_{j_2}^{m_2}(\theta_2) \Big|^2 . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \qquad (3) \end{aligned}$$

In reaction (B) (§ 2.1) two gamma-rays are emitted in cascade. In the experiments we observed the angular distribution of the 3·7 MeV gamma-rays and their angular correlation with the protons. We have extended the theory to a four-stage process, so as to be able to deal with this case. The emission of the first (0·2 MeV) gamma-ray, which is not observed, leaves the residual nucleus, ^{13}C, in an excited level with spin S_2. After the emission of the second (3·7 MeV) gamma-ray with multipolarity j_3 the residual nucleus is in the ground state with spin $S_3=\frac{1}{2}$. The angular correlation between protons emitted in the direction (θ_1, ϕ_1) and gamma-rays emitted in the direction (θ_3, ϕ_3) relative to the direction of the incident beam is given by

$$\begin{aligned} W(\theta_1, \phi_1 ; \theta_3, \phi_3)= & \sum_{M_0, M_1'', M_3} \sum_{j_2, m_2} \Big| \sum_{l_0} i^{l_0}(2l_0+1)^{1/2} \langle S_0 M_0 l_0 0 \mid JM \rangle \\ & \times \sum_{S_1, l_1} A(J, S_0, S_1, l_0, l_1) \sum_{M_1} \langle JM \mid S_1 M_1 l_1 m_1 \rangle \\ & \times Y_{l_1}^{m_1}(\theta_1, \phi_1) \langle S_1 M_1 \mid S_1' M_1' S_1'' M_1'' \rangle \\ & \times D(S_1', M_1', S_2, M_2) A(j_2) \sum_{j_3} D(S_2, M_2, S_3, M_3) \\ & \times A(j_3) \Phi_{j_3}^{m_3}(\theta_3, \phi_3) \Big|^2 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \qquad (4) \end{aligned}$$

and the angular distribution of the gamma-rays by

$$\begin{aligned} W(\theta_3)= & \sum_{M_0, M_1'', M_3} \sum_{l_1, m_1} \sum_{j_2, m_2} \Big| \sum_{l_0} i^{l_0}(2l_0+1)^{1/2} \langle S_0 M_0 l_0 0 \mid JM \rangle \\ & \times \sum_{S_1} A(J, S_0, S_1, l_0, l_1) \langle JM \mid S_1 M_1 l_1 m_1 \rangle \\ & \times \langle S_1 M_1 \mid S_1' M_1' S_1'' M_1'' \rangle D(S_1', M_1', S_2, M_2) \\ & \times A(j_2) \sum_{j_3} D(S_2, M_2, S_3, M_3) A_3(j_3) \Phi_{j_3}^{m_3}(\theta_3) \Big|^2 . \quad . \quad . \quad . \quad . \quad . \qquad (5) \end{aligned}$$

4.2. *Calculation of Angular Correlations and Distributions*

In order to calculate the angular correlations and distributions we have made use of the spins of the excited levels of ^{14}N and ^{13}C and the spin-channel factors obtained from the results of the proton distributions described in I. We have assumed further that only the lowest values of the orbital momenta of the incident and emitted particles consistent with the conservation of spin and parity have contributed to the reaction. The assignment of multipoles for the gamma-rays is discussed in § 4.3. We have combined the angular correlations, and also the angular distributions of the high energy gamma-rays in reactions (A), (B) and (C) by means of weighting factors derived from the relative yields of the different groups of protons and gamma-rays. The weighting factors were corrected for the anisotropic emission of the different particles. The results of the calculations are shown in table 1.

Table 1. Theoretical Angular Correlations and Gamma-Ray Distributions

Resonance Energy in Mev	Relative Phases of Spin-Channel Amplitudes in Reactions		Angular Correlation in		Angular Distribution of	
	(A) & (B)	(C)	Plane containing Incident Beam	Plane Perpendicular to Incident Beam	High Energy Gamma-Rays (Mixture of 3·7 and 3·9 Mev)	Low Energy Gamma-Rays (0·2 Mev)
1·13	—	—	$1+0{\cdot}037\cos(2\theta+21^\circ)+0{\cdot}012\cos(4\theta-17^\circ)$	$1+0{\cdot}24\cos^2\phi$	$1+0{\cdot}17\cos^2\theta$	$1-0{\cdot}27\cos^2\theta$
1·51	0	0	$1+0{\cdot}088\cos(2\theta-78^\circ)+0{\cdot}088\cos(4\theta-18^\circ)$	$1+0{\cdot}01\cos^2\phi$	$1-0{\cdot}24\cos^2\theta$	$1+0{\cdot}27\cos^2\theta$
	0	π	$1+0{\cdot}118\cos(2\theta-47^\circ)+0{\cdot}088\cos(4\theta-18^\circ)$	$1-0{\cdot}02\cos^2\phi$	$1-0{\cdot}19\cos^2\theta$	
	π	0	$1-0{\cdot}133\cos(2\theta+15^\circ)+0{\cdot}023\cos(4\theta-18^\circ)$	$1+0{\cdot}02\cos^2\phi$	$1-0{\cdot}35\cos^2\theta$	$1+0{\cdot}52\cos^2\theta$
	π	π	$1-0{\cdot}074\cos(2\theta+28^\circ)+0{\cdot}023\cos(4\theta-18^\circ)$	$1-0{\cdot}01\cos^2\phi$	$1-0{\cdot}31\cos^2\theta$	
1·64	0	—	$1+0{\cdot}064\cos(2\theta+20^\circ)-0{\cdot}026\cos(4\theta+25^\circ)$	$1-0{\cdot}26\cos^2\phi+0{\cdot}08\cos^4\phi$	$1+0{\cdot}19\cos^2\theta-0{\cdot}27\cos^4\theta$	$1-0{\cdot}02\cos^2\theta$
	π	—	$1-0{\cdot}095\cos(2\theta+17^\circ)+0{\cdot}050\cos(4\theta-10^\circ)$	$1-0{\cdot}17\cos^2\phi+0{\cdot}05\cos^4\phi$	$1+0{\cdot}25\cos^2\theta-0{\cdot}20\cos^4\theta$	$1-0{\cdot}03\cos^2\theta$

Magnetic dipole transition has been assumed for the 3·7 Mev gamma-rays

If we take ($\pi/2$, 0) as the direction of the protons in eqn. (1), the spherical harmonic $Y_{l_1}^{m_1}(\pi/2, 0)$ causes many of the cross terms to cancel, so that the resulting expression becomes particularly simple. But if the protons are observed at 90° to the incident beam, they will be moving in a backward direction in centre-of-mass coordinates, and then the correlation curve in the plane containing the incident beam may be substantially different from the curve for which the protons leave at 90° in centre-of-mass coordinates. If the correlation is given by

$$W(\theta_2)=1+a\cos^2\theta_2+b\cos^4\theta_2 \quad . \quad . \quad . \quad . \quad . \quad . \quad (6)$$

when $\theta_1=90^\circ$, then the change of θ_1 to $90^\circ+\alpha$ (where α is the centre-of-mass correction to θ_1) introduces two additional terms $c\sin 2\theta_2$ and $d\sin 4\theta_2$, as well as corrections to a and b which are small enough to be ignored. On rearranging the terms, the corrected correlation becomes

$$W(\theta_2)=1+\tfrac{1}{2}b+\tfrac{3}{8}a+\left[\left(\frac{a+b}{2}\right)^2+c^2\right]^{1/2}\cos(2\theta_2-y)$$
$$+\left[\left(\frac{a}{8}\right)^2+d^2\right]^{1/2}\cos(4\theta_2-x)\quad\ldots\ldots\quad(7)$$

where $\tan x=8d/a$; $\tan y=2c/(a+b)$. If a and b are approximately equal and of opposite sign, even a small value of c may alter the shape of the correlation curve considerably. The effect of the proton motion was found to be particularly important for reaction (A) at the 1·51 Mev resonance.

4.3. *Confirmation of Spins and Parities*

In paper I it was shown from the proton angular distribution experiments and from the existence of a strong 0·2 Mev gamma-ray that the 3·68 and 3·89 Mev levels in ^{13}C must have spins and parities $3/2^-$ and $5/2^+$ respectively. The observed angular distributions of the 0·2 Mev gamma-rays indicate that this gamma-ray is due to an electric dipole transition, consistent with the above assignments of spins and parities. The angular correlations and distributions of the high energy gamma-rays showed terms up to the fourth power of $\cos\theta$, agreeing with the assignment of $5/2^+$ for the level at 3·89 Mev ; gamma-rays emitted from a level with lower spin cannot give rise to terms higher than $\cos^2\theta$.

Table 2. Comparison of Experimental and Theoretical Angular Distributions of High Energy Gamma-Rays at 1·51 Mev Resonance

Experimental distribution	$1-(0{\cdot}151\pm0{\cdot}007)\cos^2\theta$
Theoretical distribution assuming magnetic dipole transition for 3·7 Mev gamma-rays	$1-0{\cdot}19\cos^2\theta$
Theoretical distribution assuming electric quadrupole transition for 3·7 Mev gamma-rays	$1-0{\cdot}29\cos^2\theta$

The experimental distribution has been obtained by the method of least squares. The theoretical distributions are in closest agreement with the experimental distribution for which the relative phase of the spin-channel amplitudes has been assumed to be 0 in reactions (A) and (B), and π in reaction (C).

The transition from the level at 3·68 Mev to the ground state might be by mixed magnetic dipole and electric quadrupole radiation, but the relative transition probabilities based on the independent particle model (Blatt and Weisskopf 1952) favour the magnetic dipole transition by 400 : 1. In the theoretical calculations we have assumed a pure magnetic dipole transition for the 3·7 Mev gamma-rays, but at the 1·51 Mev resonance we have also considered a pure electric quadrupole transition. The theoretical expression based on the magnetic dipole transition produced closer agreement with the experimental curve for the gamma-ray angular distributions (see table 2), but the influence of the 3·7 MeV gamma-rays on the distribution was much reduced by the preponderance of the 3·9 MeV gamma-rays.

Our confirmation of the assignments made in paper I for the spins and parities of the excited levels of ^{14}N is only indirect and depends on the reasonably good agreement between the theoretical curves derived by assuming these assignments and the experimental results here presented.

4.4. *Assignment of Phases*

In § 4.1 we have assumed that the amplitudes of the competing spin-channels are either in phase or 180° out of phase. It is in principle possible to determine the relative phases by comparing the theoretical and experimental curves. At the 1·51 MeV resonance the spin-channel amplitudes for the predominant reactions (A) and (B) are in phase. This is clearly shown by the angular distribution of the high energy gamma-rays and by the correlation in the plane containing the incident beam, fig. 2; it is confirmed by the angular distribution of the 0·2 MeV gamma-rays, fig. 4. The distribution of the high energy gamma-rays also suggests that in reaction (C) the spin-channel amplitudes are 180° out of phase; but we cannot make a unique assignment, as this reaction produces only 16% of the total number of counts. At the 1·64 MeV resonance the angular distribution of the high energy gamma-rays indicates that the spin-channel amplitudes in reactions (A) and (B) are 180° out of phase.

§ 5. CONCLUSION

Our results confirm the assignments made in paper I for the spins and parities of excited levels of ^{14}N and ^{13}C. The amplitudes and phases associated with the channel spins resulting from the spin orientation of the emitted proton have been calculated by K. V. Roberts (private communication) from the nuclear shell model with Russell–Saunders coupling (see Christy 1953), but agreement with our experimental values was poor. Recently, Lane (1953) has shown that the low excited states of ^{13}C are best explained by means of a nuclear shell model with intermediate coupling between the Russell–Saunders and *j-j* extremes (see also Inglis 1953).

ACKNOWLEDGMENTS

The author wishes to express his gratitude to Mr. E. S. Shire for much helpful advice and for assistance in the preparation of this paper. He wishes to thank the Atomic Energy Research Establishment, Harwell, for preparing separated isotope targets. He is indebted to Trinity College, Cambridge, for a Research Scholarship, and to the Cavendish Laboratory and the Department of Scientific and Industrial Research for financial assistance.

REFERENCES

BLATT, J. M., and WEISSKOPF, V. F., 1952, *Theoretical Nuclear Physics* (New York : Wiley).

BIEDENHARN, L. C., ARFKEN, G. B., and ROSE, M. E., 1951, *Phys. Rev.*, **83,** 586.

CHRISTY, R. F., 1953, *Phys. Rev.*, **89,** 839.

CONDON, E. U., and SHORTLEY, G. H., 1951, *The Theory of Atomic Spectra* (Cambridge : University Press).

HUTCHINSON, G. W., and SCARROTT, G. G., 1951, *Phil. Mag.*, **42,** 792.

INGLIS, D. R., 1953, *Rev. Mod. Phys.*, **25,** 390.

LANE, A. M., 1953, *Proc. Phys. Soc.* A, **66,** 977.

LITTAUER, R. M., 1950, *Rev. Sci. Instrum.*, **21,** 750.

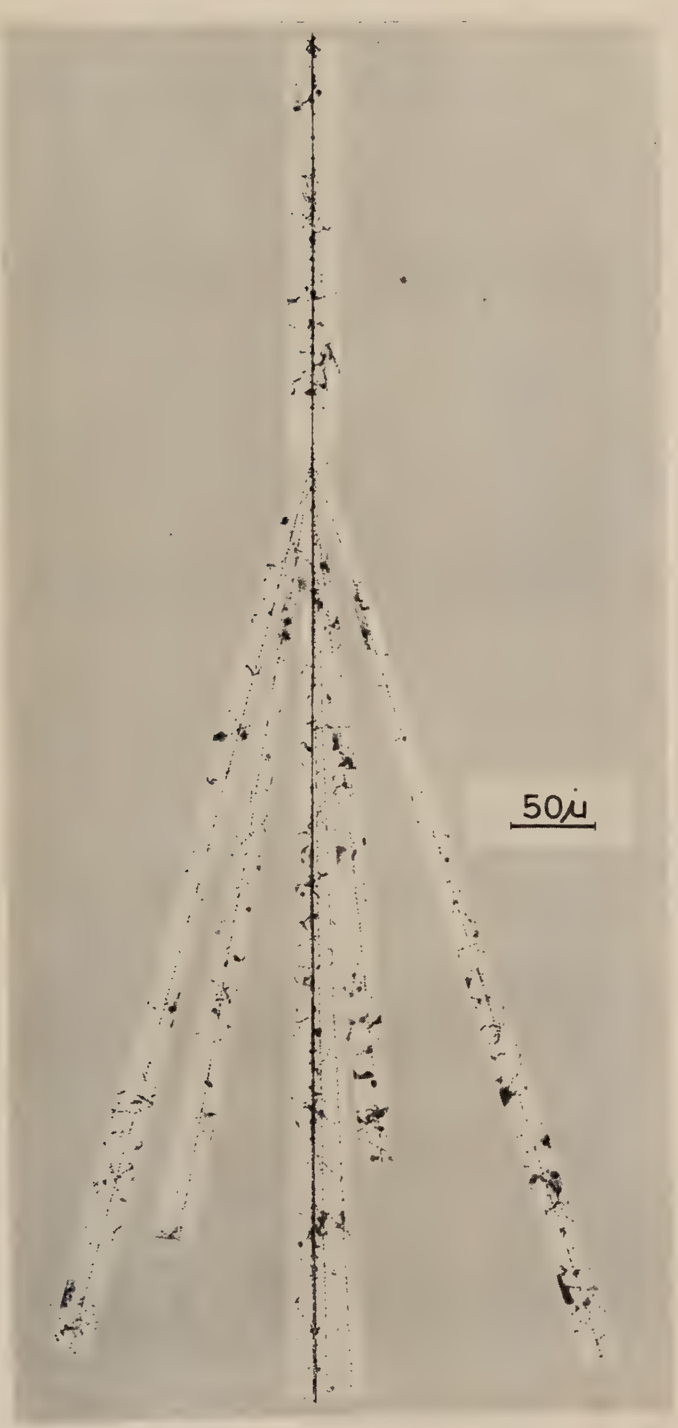
LLOYD, S. P., 1951, *Phys. Rev.*, **81,** 161.

SHIRE, E. S., WORMALD, J. R., LINDSAY-JONES, G., LUNDÉN, A., and STANLEY, A. G., 1953, *Phil. Mag.*, **44,** 1197.

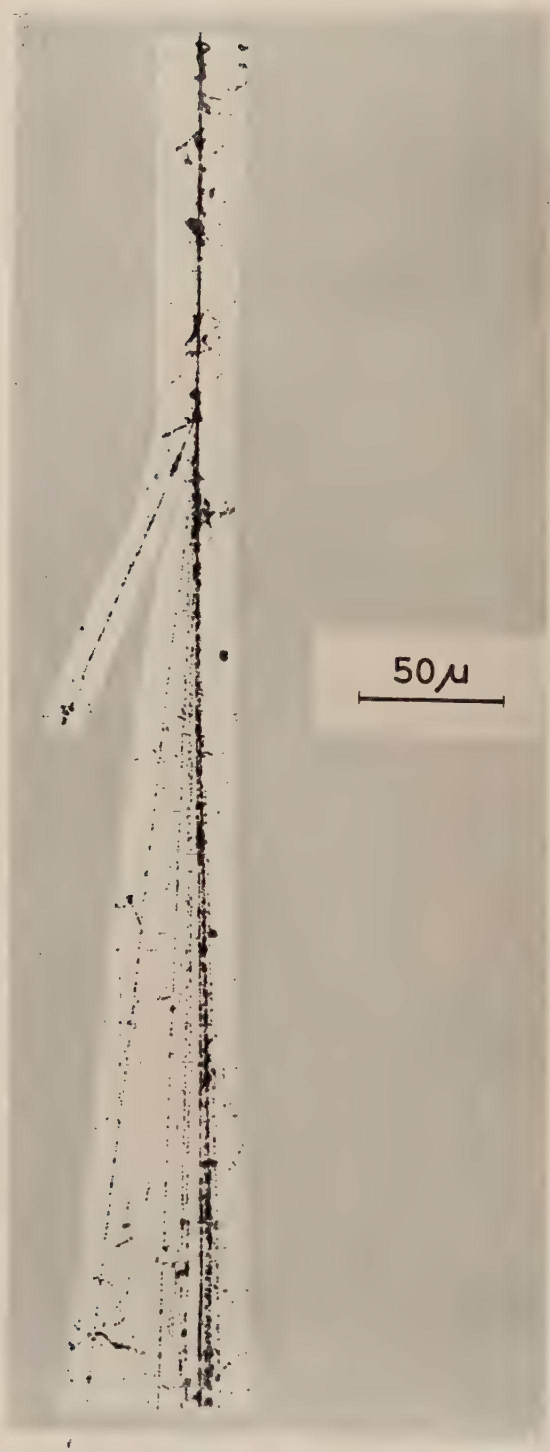
SPIERS, J. A., 1950, *Phys. Rev.*, **80,** 491.

WIGNER, E. P., and EISENBUD, L., 1947, *Phys. Rev.*, **72,** 29.

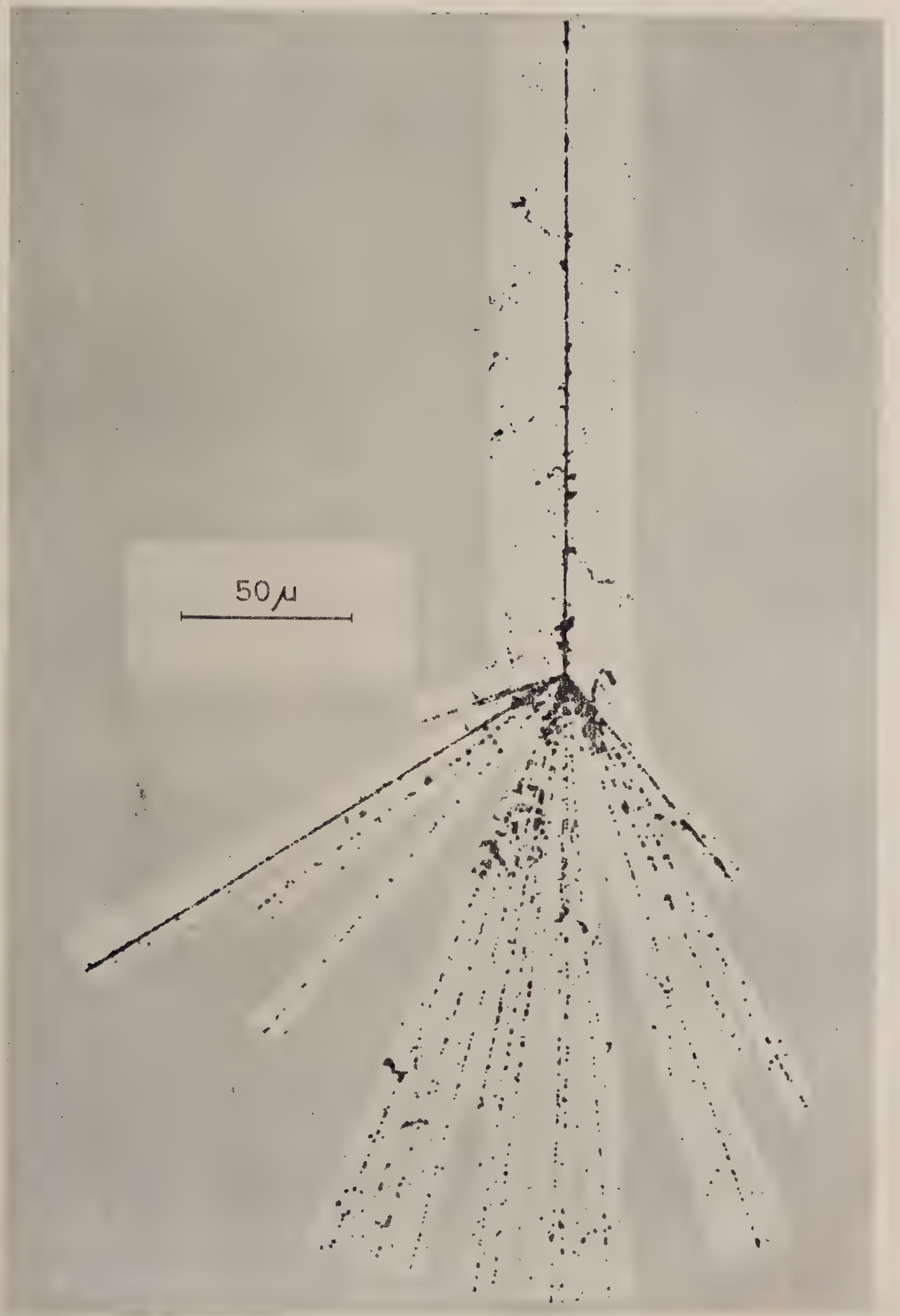
[*The Editors do not hold themselves responsible for the views expressed by their correspondents.*]



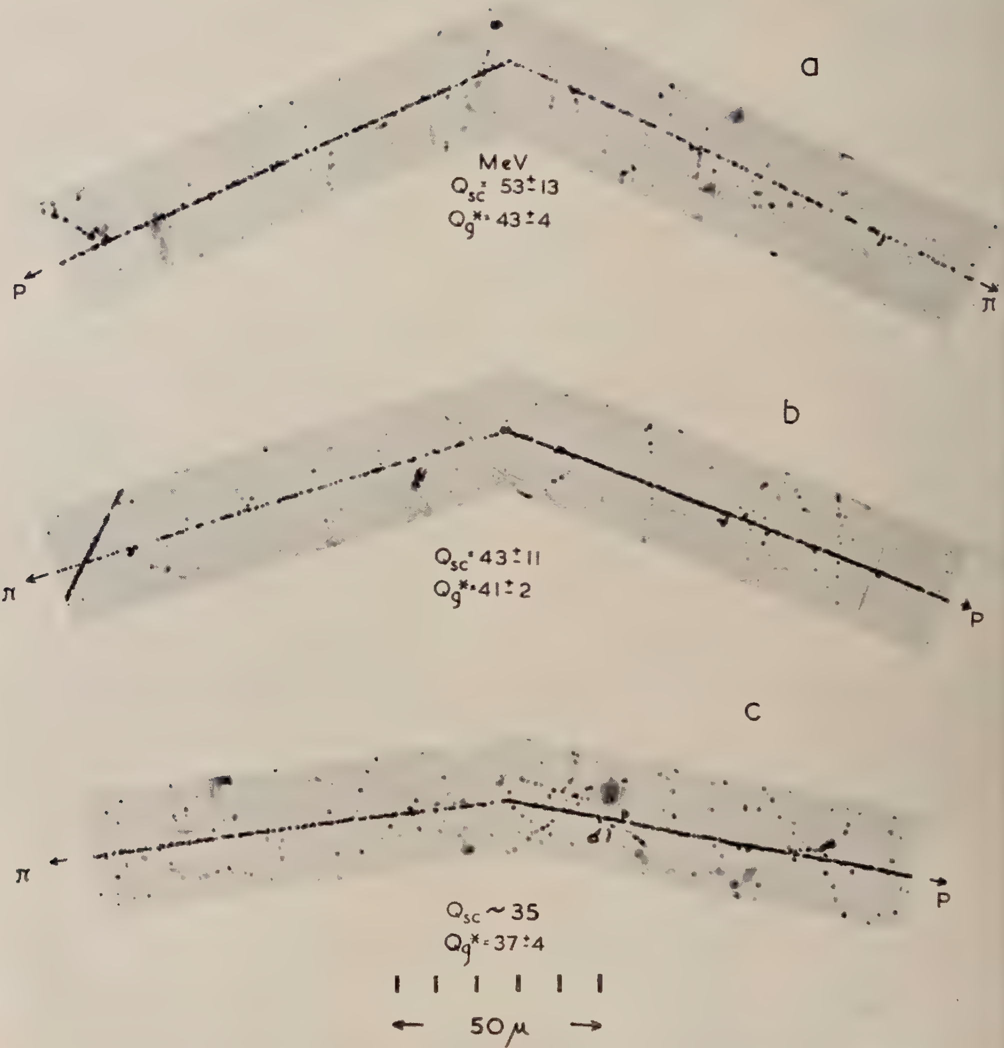
A neon nucleus ($Z=10$) is split up into a boron nucleus ($Z=5$), an α-particle (the track of which can just be distinguished beside that of the boron) and probably three protons. Some mesons are created. The struck particle may have been a hydrogen nucleus.



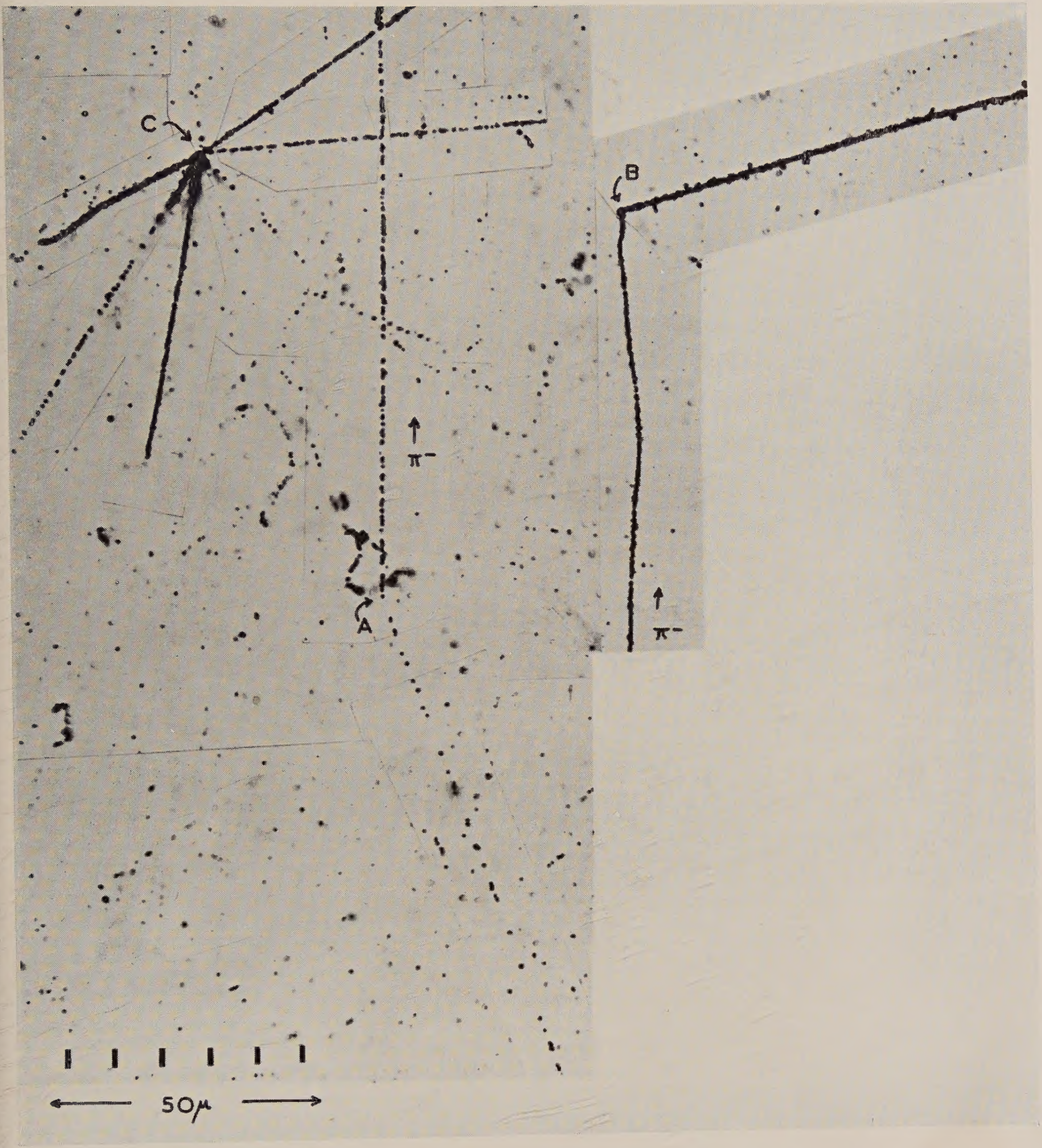
An oxygen nucleus ($Z=8$) disintegrates into two α-particles and 4 protons. Besides three ‘ thin ’ tracks and one ‘ grey ’ track are visible.



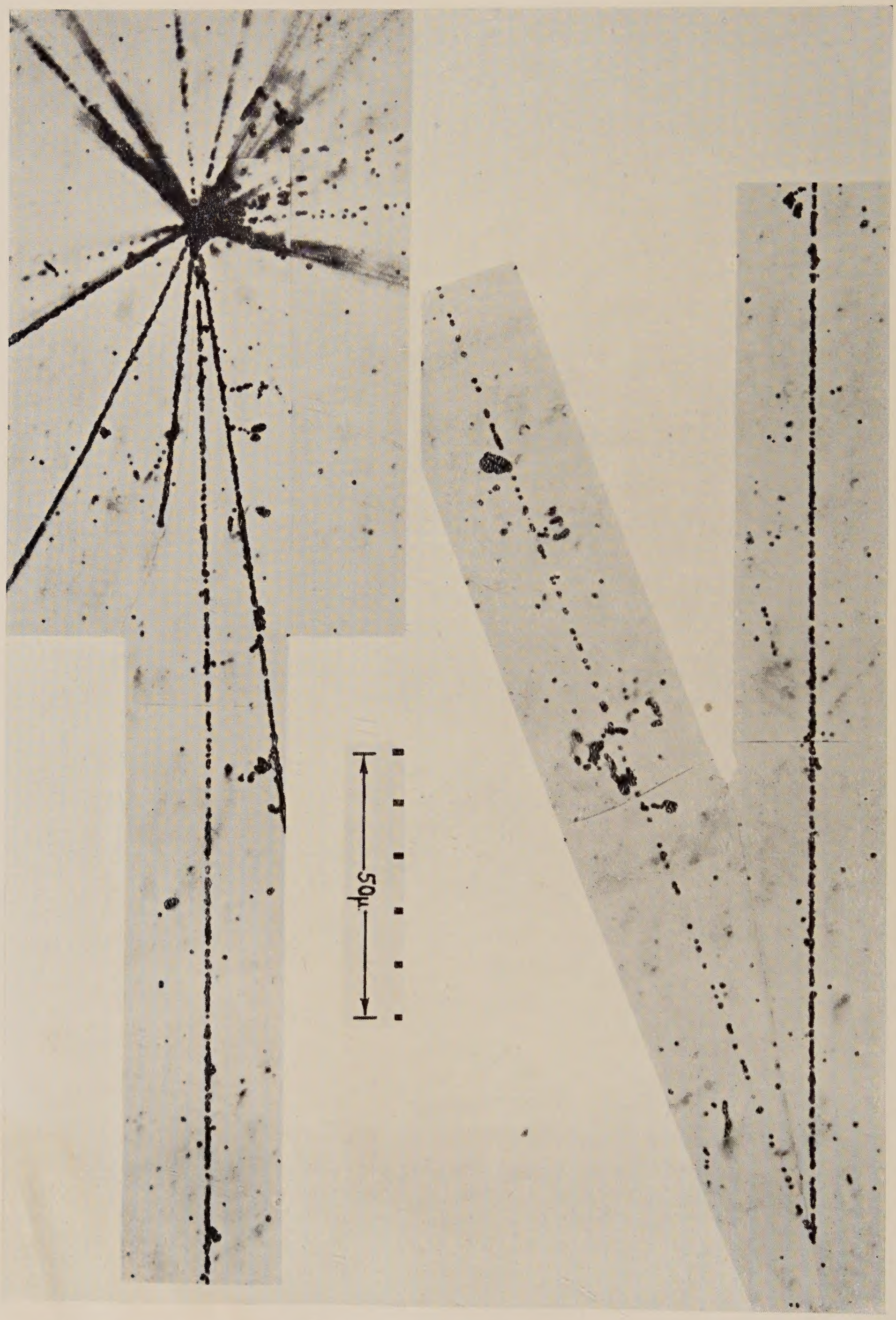
Complete disintegration of a boron nucleus. 22 tracks, 19 of which are ‘ thin ’.



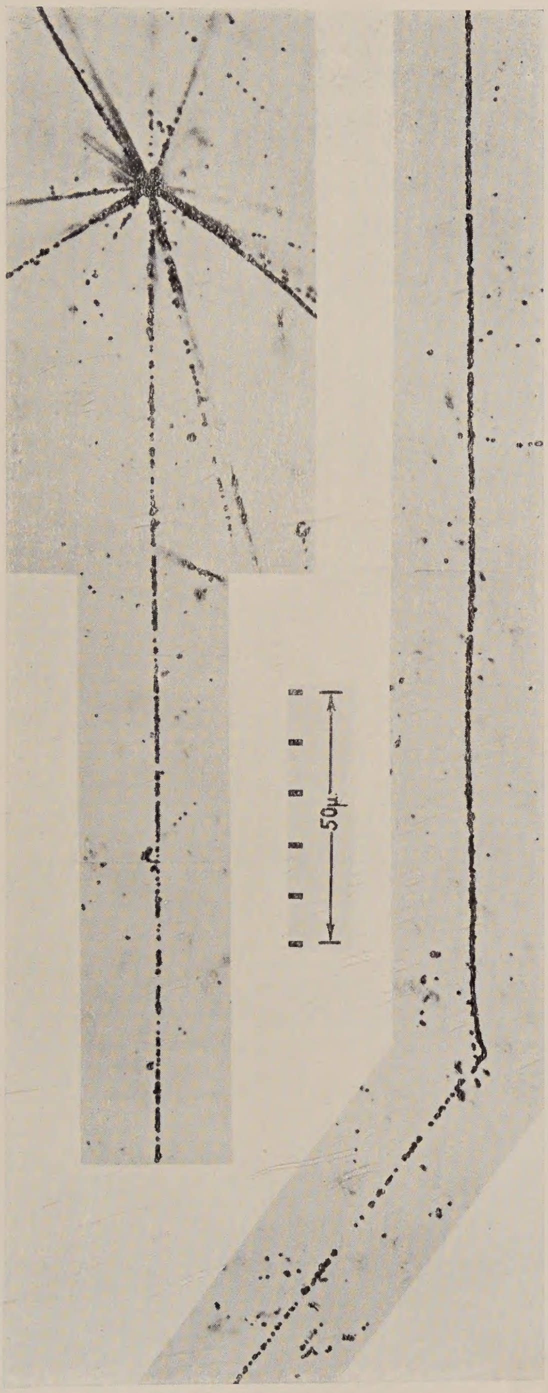
Photomicrographs (*a*), (*b*), and (*c*) show the *V*-events (1), (2), and (3) of table 1. The values Q_{Sc} are the *Q*-values given in col. 10 of Table 1 ; the values Q_{g^*} are those of col. 6 of table 2.



The event A probably represents the decay in flight of a θ^0 particle. The negative π-meson arising from the decay was arrested in the emulsion and produced a 1-prong σ-star at B. The calculated line of flight of the θ^0 particle passes through C, the centre of the disintegration $10+7p$, within the errors of measurement. The θ^0 particle was therefore probably created at C.



Event Y(B)1 : Ejected from a star of type 26+14p, the hyperon decays in flight after travelling a distance of 4·72 mm.



Event Y(B)2 : The hyperon comes from a star of type 31+3n, and reaches the end of its range after a distance of 4·64 mm. From the end of the hyperon track, there starts another track which can be identified as that of a proton, of energy 100±15 MeV.

R. H. W. JOHNSTON and C. O'CEALLAIGH Phil. Mag. Ser. 7, Vol. 45, Pl. 14.